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Fabrication details of $Ba_{1-x}K_xFe_2As_2$ films by pulsed laser deposition technique

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Abstract

Among Fe-based superconductors, potassium doped $BaFe_2As_2$ is favorable for applications because of its relatively high transition temperature and low anisotropy. To study the superconducting properties and the applicable aspects, high quality thin films of potassium doped $BaFe_2As_2$ should be fabricate. However, the high volatility of potassium makes it difficult to fabricate thin films of this compound. In this paper, we discuss the details of the experimental conditions used to fabricate $Ba_{1-x}K_xFe_2As_2$ films by *ex situ* PLD method. In the first set of samples, barium ratio in the target was controlled to make films with various potassium doping rate. However, in the second set of samples, the amount of potassium was controlled to find out optimal conditions for making high quality $Ba_{1-x}K_xFe_2As_2$ films.

Keywords: Iron-based; films; PLD

1. INTRODUCTION

Since the discovery of iron based superconductors [1], many researchers are interested in this kind of superconductors. Because of their great efforts, the transition temperature of this superconductor is raised up to 55 K [2], [3] and the unique gap symmetry has been identified [4]. Among iron based superconductors, potassium doped $BaFe_2As_2$ superconductor has small anisotropy value [5], comparable transition temperature, and high upper critical field [6] that makes it a suitable candidate for various applications [7]. In addition, the observation of peculiar gap feature in this hole doped iron based superconductor [8] makes it a more interesting material.

However, the critical current density which is one of criterions of the sample's quality, of potassium doped $BaFe_2As_2$ superconducting thin films is lower than the other iron based superconducting films [9], [10]. In spite of that, good quality film is essential in applications such as Josephson devices or 2G wire as well as in physical experiments such as vortex dynamics or reflectance.

In this paper, we present the details of experimental conditions used to fabricate potassium doped $BaFe_2As_2$ films by *ex situ* PLD method. In the first set of samples, the barium ratio was changed in the targets, and in the second set of samples the mass of potassium was controlled to find out optimal conditions. Under such conditions we successfully fabricated highly crystalline $Ba_{1-x}K_xFe_2As_2$ films.

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2. EXPERIMENT

The method of synthesizing targets is similar to that in our previous works [9], [11]. However, the molar ratio of As is decreased from as we used in previous works, so the compositional molar ratio of Fe powder (99.9%) and As pieces (99.99%) is 2.0 and 2.4, respectively. The barium lumps (99%) were used as Ba source. In the first set of samples, barium ratio in the targets was controlled to find out optimal conditions. All materials were put into alumina crucible and encapsulated in quartz ampoule, and annealed

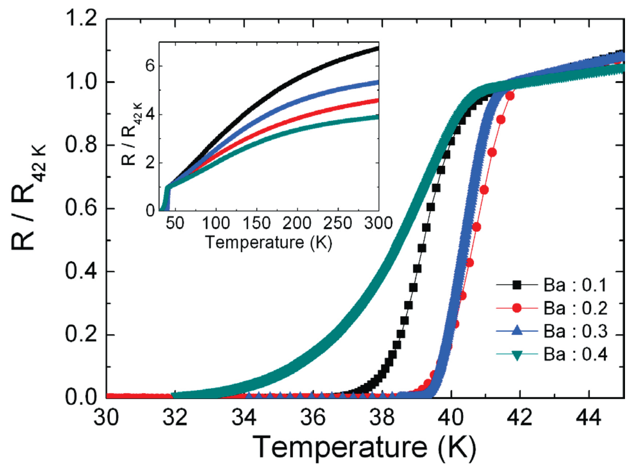

Fig. 1. Normalized resistance-temperature graph of potassium doped $BaFe_2As_2$ thin films. Resistance is normalized at 42 K. Films were fabricated by using separate targets which have different compositional ratio. Inset shows full view of the normalized resistance-temperature curves from 0 to 300 K.

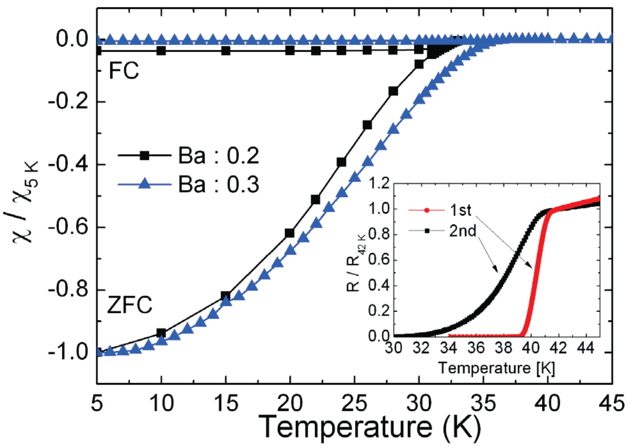

Fig. 2. Normalized magnetization-temperature curves of potassium doped $BaFe_2As_2$ thin films. Magnetization is normalized at 5 K. Films were fabricated by using targets those have different compositional ratio of Ba. One target has a compositional ratio of Ba/Fe/As = 0.2/2.0/2.4 and the other target has a ratio of Ba/Fe/As = 0.3/2.0/2.4. Inset of the figure shows the results of the repetition experiment performed on the target which has a Ba ratio of 0.3. This graph shows that the quality of the film becomes bad drastically when the second experiment was performed.

at 900 °C for 12 h. After annealing the reacted material was ground and annealed in sealed quartz ampoule again at 900 °C. The reacted compound was ground and contained in a mold of 15 mm in diameter. The contained compound was pressed into a disk and was annealed in a silica capsule at 900 °C for 8 h. The process for target fabrication was conducted in Ar (99.999%) atmosphere bag.

Precursor thin films of the synthesized targets were deposited on Al_2O_3 (*c*-cut) substrates at room temperature by means of pulsed laser deposition under the chamber's base pressure of 10^{-6} Torr. The laser energy density was 7.5 J/cm^2 at a laser flux of 400 mJ and a pulse frequency was 5 Hz. After depositing precursor thin films, they were put into a quartz ampoule along with high-purity K metal (99.5%) in Ar atmosphere. The encapsulated quartz ampoule was annealed in a furnace at 725 °C for 40 min and the heating ratio was fixed at 100 °C/hour.

The phases and the crystalline quality of the potassium-doped $BaFe_2As_2$ films were investigated by using x-ray diffractometry. The thickness and surface morphology of the films were confirmed by using scanning electron microscopy (SEM). The superconducting properties were measured by using a magnetic property measurement system (Quantum Design). The resistivity was measured by using a standard four-probe method.

3. RESULTS

Fig. 1 shows the normalized resistance versus temperature curves of hole doped $BaFe_2As_2$ films. Each film was fabricated by using separate targets that have different Ba compositional ratio of 0.1, 0.2, 0.3, and 0.4. All films were annealed at 725 °C. The high transition temperature was obtained for the films fabricated from the targets which have Ba compositional ratio of 0.2 and 0.3. These samples show similar critical temperature in resistance-temp- erature measurements. However, M-T graph, Fig. 2 shows that the films fabricated by using the target which has a Ba ratio of 0.3 exhibits Meissner effect at higher temperature while the other films doesn't show Meissner effect. For studying physical properties systematically and for the real applications, reproducibility of the samples and repetition of the experiment are very important. In this aspect, two precursor films were deposited in a row from the same target which has a Ba ratio of 0.3. Inset of Fig. 2 shows normalized resistance versus temperature curves for the potassium doped $BaFe_2As_2$ films prepared from these two precursor films. The $T_{c,zero}$ of 39 and 32 K was observed for the films which were deposited in a row, respectively. This indicates that the quality of the film becomes bad drastically when the second experiment was performed.

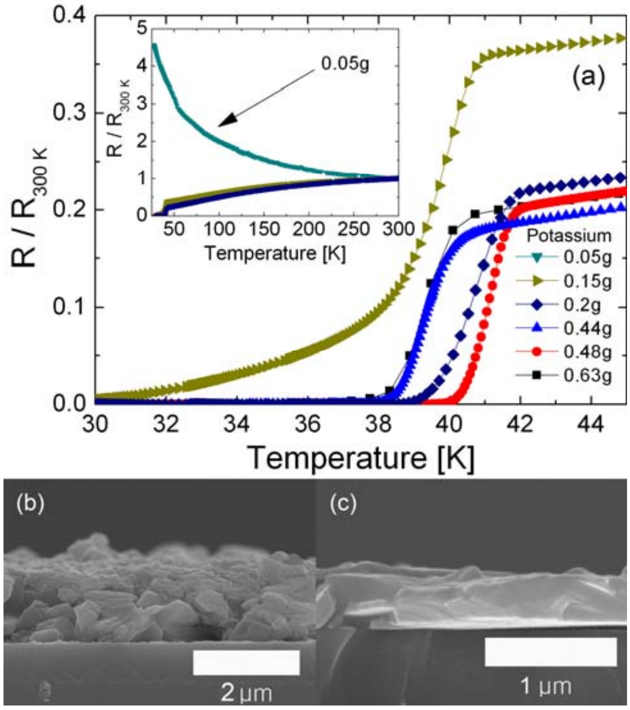

Fig. 3. (a) Normalized resistivity versus temperature graph of potassium doped $BaFe_2As_2$ thin films. Mass of potassium was controlled to find out optimal conditions. When the mass of K is 0.2 g or above similar transition temperature was obtained for the films. When K of mass 0.05 g is used, a semiconductor like behavior was noticed. The cross-sectional SEM images of the films that are annealed with K amount of (b) 0.48 g and (c) 0.2 g. The film annealed with 0.2 g of K shows smooth morphology and better grain connectivity.

This might be due to the high volatility of arsenic and the use of high density of laser energy. Therefore, the surface

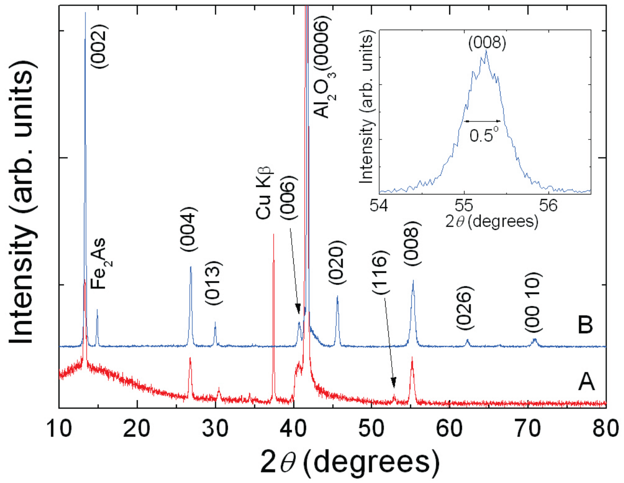

Fig. 4. X-ray diffraction θ-2θ scans. A is a sample that is annealed with K of 0.48 g and B is a sample that is annealed with K of 0.2 g. B shows higher crystallinity and one Fe_2As peak is observed at 14.8°. Inset shows the FWHM of (008) peak of sample B.

of the target must be clean before every experiment. In the second set of samples, we have varied the mass of potassium while the compositional ratio of Ba was fixed at 0.2, and all the films were annealed at 725 °C. Fig. 3(a) shows temperature versus normalized resistance of potassium doped $BaFe_2As_2$ thin films with various masses of K. When potassium of mass 0.05 g is used, semiconductor like behavior was observed in the resistance versus temperature curve. When we put K of 0.2 g or above 0.2 g with the precursor films, similar transition temperature of around 39 K was obtained for all films. It is an important to mention here that the target was cleaned after every experiment.

Fig. 3(b)–(c) show the cross-sectional SEM images of potassium doped $BaFe_2As_2$ films with different potassium amount of (b) 0.48 g and (c) 0.2 g. Thickness of these films are 1.5 μm for (b) and 450 nm for (c). The crumpled like grains were observed for the film with potassium amount of 0.48 g whereas clean and smooth morphology was observed for the film that has potassium mass of 0.2 g. This indicates that the morphology of the film which is fabricated with less amount of K of about 0.2 g is better than the film fabricated with higher amount of K. The lower amount of K enhances the crystallinity of the film as depicted in the XRD graphs in Fig.4.

Fig. 4 shows X-ray diffraction θ-2θ scans of potassium doped $BaFe_2As_2$ thin films. A and B are the curves of the films fabricated with different amount of potassium of 0.48 g and 0.2 g, respectively. B shows better crystallinity as compared to A, and the full width at half maximum of the (008) peak is 0.5° which is comparable with the iron based superconducting films fabricated by *in situ* method [11].

4. SUMMARY

Potassium doped $BaFe_2As_2$ thin films were grown by pulsed laser deposition technique. Two different set of samples were prepared, in one set barium ratio in the target was controlled whereas in other set amount of potassium was controlled to find out optimal conditions for making high quality $Ba_{1-x}K_xFe_2As_2$ films. The film fabricated by using a target which has compositional ratio of Ba/Fe/As = 0.3/2.0/2.4 has highest quality. When potassium of mass 0.2g was annealed with the precursor films, high crystalline potassium doped $BaFe_2As_2$film was obtained.

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