$Fe_{0.5}Cu_{0.5}(Ba_{1-x}Sr_x)_2YCu_2O_{7+\delta}$ (x=0, 0.5 and 1) superconductors prepared by high pressure synthesis

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Abstract

In this paper, the Fe-containing superconductors $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$, $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ were successfully prepared by common solid-state reaction followed with a procedure of high pressure synthesis. The structural change and superconducting properties in $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ ($x=0\sim 1.0$) systems were also investigated. Annealing experiments indicate that the occurrence of superconductivity in $Fe_{0.5}Cu_{0.5}(Ba_{1-x}Sr_x)_2YCu_2O_{7+\delta}$ (x=0,0.5 and 1) systems is mainly induced by the procedure of high pressure synthesis, which causes the increase of oxygen content and the redistribution of Fe atoms between Cu(1) and Cu(2) sites, but not from possible secondary phase of $YBa_2Cu_3O_{7-\delta}$, $YBaSrCu_3O_{7-\delta}$ or $YSr_2Cu_3O_{7-\delta}$ superconductors.

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I. INTRODUCTION

It is well known that $YBa_2(Cu_{1-x}M_x)_3O_{6+\delta}$ (M=Fe and Co) systems (We denote it as $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$) have been extensively investigated for providing information about the mechanism of superconductivity and understanding the correlation among superconductivity, magnetism and crystal structure in high temperature ceramic superconductors 1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16 . The main conclusions concerning its crystal structure and superconducting properties are as follows:

- a. In undoped $YBa_2Cu_3O_{7-\delta}$ superconductor, there are two different structural Cu sites: Cu(1) and Cu(2). The Cu(1) site has a square planar oxygen coordination and forms Cu-O chain, and the Cu(2) site has a five fold pyramidal coordination of oxygen and forms CuO_2 plane. There is strong evidence from neutron diffraction results that Cu(1) sites are the preferred occupation sites for Fe and Co atoms in substituted $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$ compound Cu(1) sites are Cu(1) sit
- b. Neutron and X-Ray diffraction analysis indicate that $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$ undergoes a structural phase transition from orthorhombic to tetragonal at Fe concentration $x \sim 0.12-0.15^{6,7,8}$. Electron microscopy observations indicate that the microstructure of $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$ evolves as the Fe concentration changes 9,10,11,12,13 .
- c. The superconducting transition temperature T_c , decreases with the increasing Fe concentration x, when x exceeds 0.3, superconductivity disappears completely^{1,13,14,16}.
- d. The oxygen content, $7 + \delta$, in $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$, increases with the increasing Fe concentration due to the higher valence of $Fe^{1,13,15}$.

In previous studies, several groups^{17,18,19,20,21,22,23} have reported superconductivity in $YSr_2Cu_{3-x}M_xO_{6+\delta}$ compounds with Fe (or Co) light-substitution, but the heavily-substituted compounds (x>0.3) did not exhibit superconductivity. T. Denetial²⁴ observed superconductivity with $T_c\sim 30-50{\rm K}$ in $Fe(Co~{\rm or}~Ti)$ -doped R-123 phase $YSr_2Fe_xCu_{1-x}O_{6+\delta}$ with x=0.3, while x>0.3, superconductivity could not be observed. F. Shi $et~al.^{25}$ improved the superconductivity of $YSr_2Cu_{2.7}Fe_{0.3}O_{7+\delta}$ compound by high-pressure oxygen annealing, T_c reaches 60K and a shielding fraction of nearly 100% is achieved. Recently J. Shimoyama $et~al.^{26}$ prepared $FeSr_2YCu_2O_{7+\delta}$ superconductor with $T_c\sim 60{\rm K}$ using a complex synthesis procedure and T. Mochiku $et~al.^{27}$ determined the crystal structure of this superconductor by neutron powder diffraction studies. Besides, some

Fe-containing oxides with perovskite structure, such as $(Pb_{0.5}Fe_{0.5})Sr_2(Y_{0.5}Ca_{0.5})Cu_2O_7^{28}$, $BaR(Cu_{0.5+x}Fe_{0.5-x})_2O_{5+\delta}$ $(R=Y, Sm)^{29,30,31}$, $Bi_2Sr_3Fe_2O_x^{32}$, $BaYCuFeO_5^{33}$, etc. have also been reported, but none of these compounds is superconducting.

We have prepared a Fe-containing cuprate superconductor $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ with $T_c \sim 77 \mathrm{K}$ in year 2000 by solid-state reaction and high pressure synthesis. The preliminary results of this superconductor have been reported³⁴. After that, the studies of Fe-containing superconductors were developed and new superconductors of $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ were successfully prepared. In this paper, the preparation, superconductivity, structure and the results related to these superconductors are presented.

II. EXPERIMENT

A. Sample Preparation

The predried Y_2O_3 , $BaCO_3$, $SrCO_3$, Fe_2O_3 , and CuO powders of 99.99% purity were used as the starting materials. The powders with the stoichiometric composition of $Fe_{0.5}Cu_{0.5}(Ba_{1-x}Sr_x)_2YCu_2O_{7+\delta}$ (x=0,0.5, and 1.0) were mixed, ground thoroughly and calcined twice at 925°C for 60 hours in air with intermediate grinding. The products were pressed into pellets, and calcined again at 925°C for 60 hours and cooled down to room temperature at the rate of 30°C per hour in air. These samples prepared by the common solid-state reaction procedure were labeled as AM-sample.

The AM-sample powders were oxygenated under high pressure of 6GPa at 1000° C for 0.5 hour by the addition of 5wt.% $KClO_4$ (which was used as an oxygen source) in a six-anvil of tungsten carbide high pressure apparatus. Samples were quenched from high temperature quickly by cutting off the furnace power before releasing the high pressure³⁴. These samples were labeled as HP-sample.

B. Sample Characterization

The phase and structure of these samples were characterized by powder X-Ray diffraction (XRD) analysis on an MXP18A-HF type diffractometer with Cu- K_{α} radiation. All investigated samples exhibited single phase diagram. Powder X, Finax and Rietweld programs

were used for lattice parameter calculations. The data of DC magnetization and electrical resistance were measured using a DC-SQUID magnetometer (Quantum Design MPMS 5.5T) and standard four-probe technique respectively.

III. EXPERIMENTAL RESULTS

A. The structural change and superconductivity in $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ synthesized by solid-state reaction under ambient pressure

Fig.1 shows the XRD patterns of the $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ systems of AM-samples $(x=0{\sim}1.0)$, indicating that all samples are of single phase. Fig.2 shows the changes of lattice parameters. With the increasing of Fe concentration x, lattice parameter a increases and b decreases, and a structural transition undergoes from orthorhombic to tetragonal at Fe concentration $x \sim 0.15$, afterwards lattice parameter a increases slightly. The lattice parameter c always decreases whereas unit cell volume V increases with the increasing x. Fig.3 shows some typical curves of resistivity vs. temperature, which indicates that the substitution of Fe atoms suppresses the electric conductance and superconductivity for the AM-samples, when x > 0.3, all samples become non-superconducting and exhibit semi-conducting behavior in R-T curves. The inset is the dependence of superconducting transition temperature on Fe content x, which displays a linear depression. These results are similar to those of $(Fe_xCu_{1-x})Ba_2YCu_2O_{7+\delta}$ system.

B. Superconductivity of $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ prepared by high pressure synthesis

XRD patterns of $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ HP-samples indicate that all samples (x) varies from 0 to 1.0) are still single phase after high pressure synthesis. Fig.4 shows $T_c(onset)$ vs. Fe concentration x and magnetization vs. temperature curves obtained using ZFC mode under applied external magnetic field of 10Oe, indicating that all samples exhibit superconductivity and the samples with x = 0.35-0.6 have relatively high superconducting transition temperature T_c and high superconducting volume fraction V_m . Typical curves of the dependence of resistivity and magnetization on temperature for $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$

superconductor are shown in Fig.5, it can be calculated from the ZFC and FC curves that the superconducting shielding volume fraction is 48% and the Meissner volume fraction is 31% at 10K by the relationship of $V_m = (4\pi\rho M/H)$, where ρ is the density of sample in g/cm^3 , M is mass magnetization in emu/g using ZFC and FC data respectively, and H is the applied magnetic field in Oe.

C. $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductor

As mentioned previously, the $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ sample prepared by solid-state reaction in air is semi-conductor. While after high pressure synthesis, this sample becomes superconducting. Fig.6 presents the XRD patterns of the AM-sample and HP-sample, R-T and M-T curves of HP-sample. The lattice parameters for HP-sample are a = 0.3870(3)nm, c = 1.1601(5)nm, while for AM-sample, a = 0.3871(3)nm, c = 1.1671(5)nm. From the R-T curve, the superconducting transition temperature, $T_c(onset)$ is found at 83K and $T_c(zero)$ is at 63K. From the ZFC and FC curves it can be calculated that the superconducting shielding volume fraction is 55% and the Meissner volume fraction is 22% at 10K.

D. $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ superconductor

The synthesis and crystal structure of $FeSr_2YCu_2O_{7+\delta}$ superconductor have been reported by J. Shimoyama et~al. and T. Mochiku $et~al.^{26,27}$. This suggests that $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ superconductor might be obtained. It has been known that R-123 or 1212 phase can not be prepared by solid-state reaction under ambient pressure in R-Sr-Cu-O systems. However, in $(Fe_xCu_{1-x})Sr_2YCu_2O_{7+\delta}$ system, single phase of (Cu,Fe)-1212 phase can be obtained because this phase can be stabilized by the substitution of Fe for Cu.

The $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ sample prepared under ambient pressure is not superconducting, but after high pressure synthesis, the sample becomes superconducting. This suggests that the origin of superconductivity in $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ is similar to that in $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductors of HP-samples. XRD patterns and M-T curve of $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ superconductor are shown in Fig.7, the $T_c(onset)$ is about 77K, and the sample is nearly single phase. The lattice pa-

rameters for HP-sample are a = 0.3859(4)nm, c = 1.1602(5)nm, while for AM-sample, a = 0.3871(3)nm, c = 1.1669(5)nm.

E. The annealing experiments of $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ superconductor

The oxygen content in the $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ samples prepared by solid-state reaction under ambient pressure and high pressure synthesis were determined by a volumetric method¹³. The principle of this method is to dissolve the sample in diluted hydrochloric acid, according to the liberated oxygen volume, the oxygen content is calculated. The obtained results are: $7+\delta = 7.20(2)$ in AM-sample and $7+\delta = 7.35(2)$ in HP-sample. This indicates clearly that the oxygen content is increased by high pressure synthesis.

A superconducting $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ sample with $T_c \sim 60$ K was annealed in air at 210, 290 400, 500, 700, 900 and 950°C for 2 hours step by step.

Fig.8(a) shows the magnetization vs. temperature curves of the annealed $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ HP-sample, indicating that the superconducting transition temperature T_c and superconducting volume fraction decrease rapidly with the increase of annealing temperature, and the sample became non-superconducting when annealing temperature were higher than 500°C, and even after annealed under as low as 290°C, superconductivity in this sample was almost destroyed completely. The results of the annealed $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductor are similar to these results, which are shown in Fig.8(b).

 $(46^{\circ} \sim 48^{\circ})$ Fig.9 shows the local region XRD patterns in 2θ) of the $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ AM-sample and the annealed HP-sample at different temperatures. After high pressure synthesis, the peaks of (006) and (200) shift to high angle degree and overlap each other, and with the increase of annealing temperature, the peaks shift to low angle degree again. When the annealing temperature is higher than 400°C, the positions of the peaks are nearly constant within experimental error, indicating that lattice parameters a and c stop increasing. It means that the extra oxygen introduced by high pressure synthesis is liberated almost completely at 400°C. As a result, the annealed sample becomes non-superconducting. When temperature is higher than 500°C, the main role of the annealing is the improvement of lattice distortion, homogenizing and the increase of crystal grain size, which makes the peaks narrower and makes (006) and (200) peaks gradually apart from each other.

IV. DISCUSSIONS

A well-known fact in R-123 phase superconductors is that the superconductors with higher oxygen content have smaller lattice parameters a and c. This phenomenon was also observed in the $YSr_2Cu_{2.7}Fe_{0.3}O_{7+\delta}$ superconductor²⁵ and in $FeSr_2YCu_2O_{7+\delta}$ superconductor²⁷ obtained by high oxygen pressure synthesis. In these superconductors, the diffraction peaks of (006) and (200) shift systematically to higher angle degree with the increase of oxygen content, which is similar to our results. All our HP-samples have smaller lattice parameters a and c than the corresponding AM-samples, and the lattice unit cell of HP-samples displays an obvious shrinkage. Since the relative shrinkage is not more bigger than other samples annealed under high oxygen pressure, and also for that $FeSr_2YCu_2O_{7+\delta}$ sample can be made superconducting without high pressure synthesis, thereby this shrinkage can be attributed to the increase of oxygen content, but not the pressure effect caused by the high pressure synthesis procedure. Then it can be deduced that in all $Fe_{0.5}Cu_{0.5}(Ba_{1-x}Sr_x)_2YCu_2O_{7+\delta}$ samples the oxygen content increased after high pressure synthesis, which is validated by the oxygen content measurement in $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ system where $7+\delta$ increased from 7.20(2) in AM-sample to 7.35(2) in HP-sample. The increased oxygen content provided the required charge which made the semiconducting-like AM-samples changed to superconducting HP-samples.

The diffusion of Fe atom into CuO_2 planes in R-123 systems is believed to be very unfavorable for superconductivity because of its magnetic moment, and with the increase of Fe concentration, this diffusion is inevitable in samples synthesized by common solid-state reaction³⁶. J. Shimoyama $et~al.^{26}$ used a complex annealing procedure which is believed to suppress the incorporation of Fe to the CuO_2 planes in $FeSr_2YCu_2O_{7+\delta}$ system, and finally made it superconducting with $T_c \sim 60$ K. Using neutron powder diffraction studies, T. Mochiku $et~al.^{27}$ investigated the site-mixing between Fe and Cu atoms in $FeSr_2YCu_2O_{7+\delta}$ samples synthesized through different procedures, and proved that simply increasing the oxygen content could not produce superconductivity and the ordered distribution of Fe and Cu atoms between Cu(1) and Cu(2) sites is also one of the key points to the origin of superconductivity. In our heavily Fe-doped R-1212 systems, high pressure synthesis

procedure is believed to promote the transfer of Fe atoms from Cu(2) site to Cu(1) site, and to make almost all the Fe atoms to occupy the Cu(1) chain site. This redistribution of Fe atoms is a needed requirement to ensure the occurrence of superconductivity, while the increase of oxygen content provides the needed amount of charge.

The annealing experiments of $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductors indicate clearly that the concentration of oxygen content affects superconductivity in HP-samples directly. Since annealed under the temperature as low as 290°C during a short time of 2 hours, the occupation site of Fe atoms could not be changed, but superconductivity in these superconductors was destroyed sharply. The local region XRD patterns in Fig.9 indicated that the lattice parameters increased rapidly with the increase of annealing temperature below 400°C, which suggested the release of oxygen content, and this was the main reason for the loss of superconductivity.

After the discovery of $FeSr_2YCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ superconductors, there is a doubt that the superconductivity in these superconductors is possibly from $YBa_2Cu_3O_{7-\delta}$, $YBaSrCu_3O_{7-\delta}$ or $YSr_2Cu_3O_{7-\delta}$ phase formed by phase segregation. It is known that $YSr_2Cu_3O_{7-\delta}$ superconductor with $T_c=30$ -80K can be obtained also by high pressure synthesis and its superconductivity disappears after heat treating at 300-500°C. Although the single phase of XRD pattern results and high superconducting volume fraction of these superconductors have excluded these disputes, the achievement of $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductor and the similarity of its superconductivity to those of $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ superconductors eliminate this doubt. The annealing experiments also eliminate the possibility that the superconductivity in $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ HP-sample is from $YBa_2Cu_3O_{7-\delta}$ phase due to phase segregation, since it is well known that the superconductivity in $YBa_2Cu_3O_{7-\delta}$ can not be destroyed by annealing at the temperature of 200-500°C and the T_c of $YBa_2Cu_3O_{7-\delta}$ obtained by high pressure synthesis is $92K^{35}$.

The discovery of these heavily-substituted superconductors with magnetic metal of Fe atoms provides new platforms to investigate the mechanism of high temperature superconductivity and the correlation of magnetism and superconductivity, and it seems that we need to reconsider the influence of magnetism on superconductivity.

V. CONCLUSIONS

Three kind of superconductors with high Fe concentration $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$, $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ and $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$ were successfully prepared by solid-state reaction and high pressure synthesis. The high pressure synthesis resulted in the increase of oxygen content and redistribution of Fe and Cu atoms between Cu(1) and Cu(2) sites, which are two key factors for the occurrence of superconductivity. The annealing experimental results indicate these clearly and eliminate the doubt of superconductivity relative to possible secondary phase of $YBa_2Cu_3O_{7-\delta}$, $YBaSrCu_3O_{7-\delta}$ or $YSr_2Cu_3O_{7-\delta}$ superconductors.

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J.M. Tarascon, P. Barboux, P.F. Miceli, L.H. Greene, G.W. Hull, M. Eibschutz and S.A. Sunshine, Phys. Rev. B. 37, 7458(1988)

² V.N. Narozhnyi and V.N. Kochetkov, Phys. Rev. B. **53**, 5856(1996)

³ P. Bordet, J. L. Hodeau, P. Strobel, M. Marezio and A. Santoro, Solid State Comm. 66, 435(1988)

⁴ T. Kajitani, K. Kusaba, M. Kikuchi, Y. Syono and M. Hirabayashi, Japn. J. Appl. Phys. 27, L354(1988)

⁵ T. Kajitani, K. Kusaba, M. Kikuchi, Y. Syono and M. Hirabayashi, Japn. J. Appl. Phys. 26, L1727(1987)

⁶ X.S. Wu, S.S. Jiang, C.C. Lam, D.W. Wang, X.R. Huang, Z.H. Wu, Y. Xuan and X. Jin, Phys. Stat. Sol. (a) **157**, 439(1996)

⁷ X.S. Wu, C. Gou, C. C. Lam, W. M. Chen, D. F. Chen, K. Sun, W. Ji, W. J. Liu, X. Jin and S. S. Jiang, Physica C 282, 833(1997)

⁸ H. Obara, H. Oyanagi, K. Murata, H. Yamasaki, H. Ihara, M. Tokumoto, Y. Nishihara and Y. Kimura, Japn. J. Appl. Phys. 27, L603(1988)

⁹ H. Renevier, J.L. Hodeau, M. Marezio and A. Santoro, Physica C **220**, 143(1994)

¹⁰ G. V. S. Sastray, R. Wordenweber and H.C. Freyhardt, J. Appl. Phys. **65**, 3975(1989)

¹¹ Z. Hiroi, M. Takano, Y. Takeda, R. Kanno and Y. Bando, Japn. J. Appl. Phys. **27**, L580(1988)

- W.M. Chen, X.X. Yao, Yuan Chang Guo, Hua Kun Liu and S.X. Dou, J of Superconductivity: Incorporating Novel Magnetism 13, 129(2000)
- ¹³ Zhang Jincang, Liu lihua, Li Jianqi, Chen Dong, Li Xigui and Cheng Guosheng, Phys. Rev. B.
 65, 054513(2002)
- Y. Oda, H. Fujita, H. Toyoda, T. kaneko, T. Kohara, I. Nakada and K. Asayama, Japn. J. Appl. Phys. 26, L1660(1987)
- ¹⁵ J.F. Bringley, T.M. Chen, B.A. Averill, K.M. Wong and S.J. poon, Phys. Rev. B. **38**, 2432(1988)
- ¹⁶ S.K. Nikogosyan, A.A. Sahakyan, H.N. Yeritsyan, V.A. Grigoryan, E.G. Zargaryan, A.G. Sarkissyan, Physica C 299, 65(1998)
- $^{17}\,$ P.R. Slater and C. Greaves, Physica C $\mathbf{180},\,299$ (1991)
- ¹⁸ B. Okai, Japn. J. Appl. Phys. **29**, L2180 (1990)
- ¹⁹ S. A. Sunshine, L.F. Schneemeyer, T. Siegrist, D.C. Douglass, J.V. Waszczak, R.J. Cava, E.M. Gyorgy and D.W. Murphy, Chem. Matter 1, 331 (1989)
- ²⁰ Q. Xiong, Y. Y. Xue, J. W. Chu, Y. Y. Sun, Y. Q. Wang, P. H. Hor and C. W. Chu, Phys. Rev. B. 47, 11337 (1993)
- $^{21}\,$ M. G. Smith, R. D. Taylor and J. D. Thompson, Physica C $208,\,91$ (1993)
- ²² M. Pissas, G. Kallias, E. Moraitakis, D. Niarchos and A. Simopoulous, Physica C **234**, 127(1994)
- V. Terziev, R. Suryanarayanan, Mamidanna S. R. Rao, L. Ouhammou, O. Gorochov, and J. L. Dorman, Phys. Rev. B. 48, 13037(1993)
- ²⁴ Tohru Den and Tamaki Kobayashi, Physica C **196**, 141(1992)
- ²⁵ F. Shi, W.J. Bresser, M. Zhang, Y. Wu, D. McDaniel and P. Boolchand, Phys. Rev. B. 54, 6776(1996)
- $^{26}\,$ J. Shimoyama, K. Otzschi, T. Hinouchi and K. Kishio, Physica C $\bf 341\text{-}348,\,563(2000)$
- ²⁷ T. Mochiku, Y. Mihara, Y. Hata, S. Kamisawa, M. furuyama, J. Suzuki, K. Kadowaki, N. Metoki, H. Fujii and K. Hirata, J. of Phys. Soc. Japan. 71, 790(2002)
- T. Maeda, M. Taniwaki, K. Isawa, K. Sakuyama and H. Yamauchi, Advances in superconductivity V, proceedings of the 5th international symposium on superconductivity (ISS'92) November 16-19, 1992, Kobe, Eds. Bando Vamauchi
- ²⁹ J. Nakamura, J. Linden, H. Suematsu, M. Karppinen and H. Yamauchi, Physica C 338, 121(2000)
- M. Nagase, J. Linden, H. Suematsu, M. Karppinen, H. Yamauchi, Phys. Rev. B. 59, 1377(1999)

- ³¹ P. Karen, P.H. Andersen and A. Kjekshus, J. Solid State Chem. **101**, 48(1992)
- ³² V. Sedykh, I. S. Smirnova, E. V. Suvorov, A. V. Dubovitskii and V. I. Kulakov, Physica C 336, 239(2000)
- ³³ L. Er-Rakho, C. Michel, Ph. Lacorre, and B. Raveau, J. Solid State Chem. **73**, 53191988)
- ³⁴ Z. A. Ren, G.C. Che, H. Xiong, K.Q. Li, Y.S. Yao, D.N. Zheng, Y.M. Ni, S.L. Jia, H. Chen, C. Dong, J.L. Shen and Z.X. Zhao, Solid State Comm. **119**, 579(2000)
- $^{35}\,$ Bin Okai and Masatsune Onta, Japn. J. of Appl. Phys. ${\bf 1378},\,30(1991)$
- $^{36}\,$ E. Suard, I. Mirebeau, V. Caignaert, P. Imbert and A. M. Balagurov, Physica C $\bf 288,\,10(1997)$

Figure Captions

- Fig.1 Typical XRD patterns for AM-samples of $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ system
- Fig.2 Lattice parameters a, b, c and unit cell volume V vs. x in $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ system for AM-samples
- Fig.3 Typical R-T curves and T_c vs. x in $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ system for AM-samples
- Fig.4 M-T curves obtained using ZFC mode under the applied field of 10Oe for HP-samples of $(Fe_xCu_{1-x})BaSrYCu_2O_{7+\delta}$ system; insert shows T_c vs. x
- Fig.5 Typical R-T and M-T curves under the applied field of 10Oe for $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ HP-sample superconductor
- Fig.6 XRD patterns of AM-sample and HP-sample and R-T and M-T (applied field of 10Oe) curves of HP-sample of $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$
- Fig.7 XRD patterns of AM-sample and HP-sample and M-T curves (applied field of 10Oe) of HP-sample of $Fe_{0.5}Cu_{0.5}Sr_2YCu_2O_{7+\delta}$
- Fig.8 (a) M-T curves obtained using ZFC mode under applied field of 10Oe for $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ superconductor annealed at different temperatures (b) For $Fe_{0.5}Cu_{0.5}Ba_2YCu_2O_{7+\delta}$ superconductor annealed at different temperatures
- Fig.9 Local region XRD patterns of $Fe_{0.5}Cu_{0.5}BaSrYCu_2O_{7+\delta}$ AM-sample and HP-sample annealed at different temperatures

















