Ag Doping Effects in FeSe_{0.5}Te_{0.5} superconductorM. Migita^{1*}, K. Sugai¹, M. Takeda¹, M. Uehara¹, T. Kuramoto¹,
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Ag added bulk FeSe_{0.5}Te_{0.5} single crystal like samples Ag_x(FeSe_{0.5}Te_{0.5})_{1-x} were prepared by solid-state reaction. The x , nominal concentration of Ag, was ranged between 0 and 8%. The pure Ag or Ag compound was not observed in the X-ray diffraction patterns of Ag doped samples. The temperature and magnetic field dependences of magnetizations of prepared samples were measured using a superconducting quantum interference device (SQUID) magnetometer. The T_c was about 14K up to $x=4\%$, but the T_c decreased gradually to 11K above $x=5\%$. The calculated lattice parameter a is approximately constant with Ag addition. On the other hand, the c is nearly constant in the sample of $x \leq 5\%$, but it decreases rapidly in $x \geq 6\%$. The x dependence of T_c corresponds to the lattice parameter c . It means that, Ag might replace Fe and partially distorts a tetragonal FeSe_{0.5}Te_{0.5} crystal structure in $x > 6\%$. The magnetization decreased with increasing of x , but the fishtail-like humps were observed in the magnetization curves. The maximal $J_c(x)/J_c(0)$ was about 1.5 at $x=5\%$ and 5T under the applied magnetic field H perpendicular to the c -axis. The pinning potentials (U_0) were estimated from relaxation measurement of the magnetization at 5K when H was parallel to the c -axis. As a result, the sample of $x=5\%$ is considered to be more susceptible to flux creep than the sample of $x=0\%$.

Key words: Critical current density, Critical state model, Magnetization, FeSe_{0.5}Te_{0.5}, Ag, Magnetic relaxation

1. INTRODUCTION

Since the discovery of high temperature superconductivity LaFeAsO_{1-x}F_x with a critical temperature T_c of 26K [1], extensive research has been performed on FeAs-based superconductivity, and various kinds of superconductors have been discovered. However, fundamental research and potential applications of these materials may be limited by the presence of the poisonous element arsenic. Thus, discovery of As-free FeSe with a T_c of 8K which has the simplest structure among all iron-based superconductors [2] made basic research of iron-based superconductor active. The T_c of FeSe increases up to about 15K by partial substitution of selenium with tellurium or sulfur [3-8]. Furthermore, T_c can be raised dramatically by applying pressure, reaching 27K in FeSe_{0.82} at 1.48 GPa [3], which indicates an important role for the pressure in the superconductivity of the 11 system. Ion substitution is a convenient technique for generating effective internal chemical pressure, which has been exploited widely in the some superconductors [9-10]. Although there are a lot of reports that superconductive properties were improved by adding metallic elements in a superconductor matrix [11-13], there are few detailed reports which doped Ag to FeSe_{0.5}Te_{0.5}.

In this paper, we investigated the influence on the pinning property by adding Ag-powder in FeSe_{0.5}Te_{0.5} superconductor, which were studied by the magnetization analysis, using the critical state model.

2. Sample preparation

The Ag/FeSe_{0.5}Te_{0.5} samples were prepared by solid-state reaction. Powders of Fe, Se, Te and Ag were grounded and mixed with the mole ratio of 1- x : 0.5(1- x) : 0.5(1- x) : x , where x was the nominal concentration of Ag from 0% to 8%. The fully mixed powder was first pushed into an alumina tube and sealed in an evacuated quartz tube that was heated at 1323K for 20h. Subsequently, it was cooled to 673K and annealed for 200h to stabilize the superconductive layers. The prepared samples were characterized by means of powder X-ray diffraction (XRD) using Cu-K α radiation, and energy dispersive X-ray spectroscopy (EDAX). The temperature and magnetic field dependences of magnetizations were measured for the prepared single crystal like samples with typical size of $3.5 \times 3.4 \times 1.4$ (c -axis) mm³, using a superconducting quantum interference device (SQUID) magnetometer.

3. Results and discussions

In Fig. 1, the XRD patterns are shown for Ag/FeSe_{0.5}Te_{0.5} samples of $x=0\%$, 2%, 5% and 8%. As shown in this figure, most of the peaks were well indexed using a space group of $P4/nmm$ for the tetragonal FeSe_{0.5}Te_{0.5} superconducting phase. The small additive peaks corresponded to FeTe₂ and Se-rich tetragonal FeSe_{0.5+ δ} Te_{0.5- δ} impurities. Though the existence of Ag₂Te was observed by the EDAX analysis in $x=5\%$ sample, the XRD peaks corresponding to Ag or Ag compound were not observed.

Fig. 1 The XRD patterns of $x=0\%$, 2% , 5% and 8% . The impurity peaks of FeTe_2 is observed in $x=0$ sample. The impurity peaks of Se-rich appear on the right-hand side of the peaks of $\text{FeSe}_{0.5}\text{Te}_{0.5}$.

The calculated lattice parameter a and c in the samples of $x=0\%$ to 8% were shown in Fig. 2. The a is approximately constant with Ag addition. On the other hand, the c is nearly constant in the sample of $x \leq 5\%$, but it decreases rapidly in $x \geq 6\%$. It can be thought that these changes in the lattice parameters show an occurrence of chemical pressure by partial distortion of crystal structure.

Fig. 2 x -dependence of lattice parameters a and c .

In Figs. 3 and 4, the TEM images of ab -plane are shown for $\text{Ag}/\text{FeSe}_{0.5}\text{Te}_{0.5}$ samples of $x=0\%$ and 5% , respectively. The diffraction pattern of $[001]$ corresponds to tetragonal crystal one. The lattice parameter a is about $3.8\text{\AA}$. This accords the result shown in Fig. 2.

Fig. 5 shows the zero-field cooled (ZFC) and field cooled (FC) direct current (DC) magnetic susceptibilities under an external magnetic field of 1mT for the samples of $x=0\%$, 4% , 5% and 6% , where the magnetic field H was applied perpendicular to the c -axis of the sample. The FC and ZFC signals were very small above $x=6\%$. The x dependence of T_c was shown in the inset of Fig. 5. The T_c is about 14K up to $x=4\%$, but the T_c decreases gradually to 11K above $x=5\%$. The x dependence of T_c corresponds to the lattice parameter c in Fig. 2. It seems that rapid shrinkage of the c by Ag doping degrades superconductivity.

Figs 6 and 7 show the magnetization hysteresis loops and estimated J_c by Bean's model [14] at 5K for $x=0\%$ to 6% . The magnetic fields applied perpendicular and parallel to the c -axis, respectively. In this case, the magnetization of unknown ferromagnetic impurities observed at 15K is subtracted from the original data. As shown in Figs. 6 and 7 of magnetization curves, the remarkable fishtail-like humps were observed for $x=5\%$. Especially, magnetization and J_c under the magnetic fields perpendicular to the c -axis is larger than that of $x=0\%$ above 2T .

Fig. 3 TEM image of ab -plane of $x=0\%$. The lower left inset is an enlargement of a real image, and the upper right inset shows the diffraction pattern of $[001]$. These results correspond to tetragonal crystal.

Fig. 4 TEM image of ab -plane of $x=5\%$. The lower left inset is an enlargement of a real image, and the upper right inset shows the diffraction pattern of $[001]$. These results correspond to tetragonal crystal.

Fig. 5 DC magnetic susceptibilities at 1mT. x dependence of T_c was shown in the inset.

Fig. 6 M - H curves and estimated J_c under the magnetic fields perpendicular to the c -axis.

Fig. 7 M - H curves and estimated J_c under the magnetic fields parallel to the c -axis.

The x dependence of $J_c(x)/J_c(0)$ under the magnetic fields perpendicular and parallel to the c -axis are shown in Figs. 8 and 9, where $J_c(0)$ is the J_c of $x=0\%$ sample. As shown in these figures, the J_c decreases with Ag addition up to $x=4\%$. It seems that the pinning efficiency decreases, because Ag_2Te was bulked as Ag doping. On the other hands, the enhancement of J_c can be observed at $x=5\%$ under the magnetic fields perpendicular to the c -axis, and the maximal $J_c(x)/J_c(0)$ is about 1.5 at 5T. These results may show that though the antiferromagnetic FeTe_2 and paramagnetic Ag_2Te may act as the normal point pinning centers, weakly superconducting Se-rich $\text{FeSe}_{0.5+\delta}\text{Te}_{0.5-\delta}$ phase should be a dominant $\Delta\kappa$ pinning center in this system. The deterioration of superconductivity occurs above $x=6\%$.

Fig. 8 x dependence of $J_c(x)/J_c(0)$ under the magnetic field perpendicular to the c -axis.

Fig. 9 x dependence of $J_c(x)/J_c(0)$ under the magnetic field parallel to the c -axis.

The relation of the relaxation of the magnetization to time t under a magnetic field parallel to the c -axis at 5K for $x=0\%$ and 5% are shown in Fig. 10 and Fig. 11, respectively. Here, M_0 is M at $t=0$. Generally, the logarithmic relaxation rate of magnetization vs. t is proportional to pinning potential U_0 .

The magnetic field dependence of U_0 at 5K for $x=0\%$ and 5% samples which were estimated by logarithmic relaxation rate of magnetization is shown in Fig. 12. The estimated pinning potential of our result at 5K were up to 24meV. These results were slightly smaller than other similar works for YBCO or Bi-2223 [15-17]. For the $x=0\%$ sample in $\mu_0H \geq 1000\text{mT}$, the U_0 increases with the increasing magnetic field, and it corresponds to the fishtail-like humps of $x=0\%$ sample in Fig. 7. On the other hand, although the fishtail-like humps of $x=5\%$ sample was observed in Fig. 7, the U_0 of $x=5\%$ sample

in Fig. 12 monotonously decreases with the increasing magnetic field, and it is contradictory to the fishtail-like humps of $x=5\%$ sample in Fig. 7. The sample of $x=5\%$ is considered to be more susceptible to flux creep than the sample of $x=0\%$. The pinning potential of the superconductors is sensitive to changes in the flux density due to the flux creep. Therefore, the future consideration will be needed to accurately estimate the U_0 from the logarithmic relaxation rate of magnetization of the superconductor, including the $\Delta\kappa$ pinning.

Fig. 10 The relaxation of the magnetization at $x=0\%$ under the magnetic field parallel to the c -axis at 5K.

Fig. 11 The relaxation of the magnetization at $x=5\%$ under the magnetic field parallel to the c -axis at 5K.

Fig. 12 The U_0 of $x=0\%$ and 5% samples under a magnetic fields parallel to the c -axis at 5K.

4. Conclusion

The $\text{Ag}_x(\text{FeSe}_{0.5}\text{Te}_{0.5})_{1-x}$ samples were prepared by solid-state reaction. The result of XRD and EDAX showed that the Ag was not doped into the lattice site of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ but the Ag_2Te was formed. The calculated lattice parameter a was approximately constant with Ag addition. On the other hand, the c is nearly constant in the sample of $x \leq 5\%$, but it decreases rapidly in $x \geq 6\%$. It was thought that these changes in the lattice parameters showed an occurrence of chemical pressure by partial distortion of crystal structure. It is needed micro-structural analysis for the present system by TEM measurement, which shall be done in the near future.

The T_c was about 14K in $x \leq 4\%$, but it gradually decreased to 11K in $x \geq 5\%$. Superconductivity became ambiguous at 5K for the samples in $x \geq 6\%$. These results are corresponding to the decreasing of the lattice parameter c . It seems that rapid shrinkage of the lattice parameter c of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ by Ag doping degrades superconductivity.

The remarkable fishtail-like humps were observed in the magnetization curves of $x=5\%$ sample under the applied magnetic field perpendicular and parallel to the c -axis. Especially, the magnetic hysteresis of $x=5\%$ sample under the applied magnetic field perpendicular to the c -axis is larger than that of $x=0\%$ sample above 2T. The maximal $J_c(x)/J_c(0)$ is about 1.5 in $x=5\%$ sample at 5T. These results may show that the Se-rich impurities efficiently act as the $\Delta\kappa$ pinning center at $x=5\%$.

The pinning potential U_0 was evaluated from the logarithmic relaxation rate of magnetization relaxation measurement. As a result, the sample of $x=5\%$ is considered to be more susceptible to flux creep than the sample of $x=0\%$. The accurate estimation of U_0 of superconductors including $\Delta\kappa$ pinning centers from the magnetic relaxation is a challengeable problem in the future.

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