Focused ion beam microfabrication of single-crystal nanobridge toward Fe(Te,Se)-based Josephson device

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Abstract. We report the fabrication and the transport measurements of FeTe₁ₓSeₓ (ₓ=0.4, 1) nanobridges along the c axis, toward the appearance of Josephson effects in single-crystal devices. Cross sectional areas of both FeTe₀.₄Se₀.₄ and FeSe nanobridges were systematically reduced to 0.06 µm² by using a new method based on the focused ion beam (FIB) techniques. The critical current Ic measured by the current-voltage characteristics is roughly two orders of magnitude smaller than that for the conventional microbridges with larger cross sections, while the IcRn product, where Rn is the normal-state resistance along the c axis, is still 3-4 times larger than the theoretical value for the appearance of Josephson effects. We argue the importance of the development of single-crystal Josephson devices and the comparison between FeTe₀.₄Se₀.₄ and FeSe nanobridges.

1. Introduction
Iron chalcogenide (FeCh) superconductors have attracted much attention because novel phenomena were discovered in spite of the simple crystal structure composed of only FeTe/Se layers. Although the superconducting transition temperature Tc in FeSe and FeTe₁ₓSeₓ is only 9 K [1] and 14 K [2], respectively, it can be remarkably enhanced by applying pressure [3] and intercalating spacer layers [4]. In addition, an FeSe monolayer grown on a SrTiO₃ substrate shows a large Tc ~65 K [5]. The extremely small Fermi energy observed in FeSe implies a close access to the BCS-BEC (Bardeen-Cooper-Schrieffer to Bose-Einstein-Condensate) crossover region [6,7]. More interestingly, a topological surface superconductivity [8,9] and a possible Majorana bound state [10,11] have been observed in FeTe₁ₓSeₓ, suggesting that this compound is a new candidate for topological quantum matter. Thus, the Josephson effects become more important as a key probe to derive unique properties of FeCh superconductors and develop applications toward future quantum technology. In particular, the development of single-crystal Josephson devices is crucially required to observe the topological quantum properties.

Recently, we reported the achievement of a departing current limit in the c axis microbridges of FeTe₁ₓSeₓ single crystals [12]. This demonstrates that the departing current density determined by a process where the kinetic energy of supercurrent exceeds the superconducting condensation energy can be explored by the transport measurements of the single-crystal devices fabricated by the focused ion beam (FIB) techniques. We also confirmed that FeCh superconductors did not show any sign of the
intrinsic Josephson effect, in contrast to other iron-based superconductors such as (V$_2$Sr$_2$O$_6$)Fe$_2$As$_2$, where the superconducting layers are effectively decoupled by the insulating (VSr$_2$O$_3$) layers [13]. The observation of the conventional Josephson effects has been reported for the in-plane nanobridges fabricated from FeTe$_{0.4}$Se$_{0.5}$ thin films [14].

In this study, we successfully fabricate the $c$-axis nanobridges made of FeTe$_{1.4}$Se$_{2.4}$ ($x=0.4, 1$) single crystals by using the FIB techniques. We systematically reduce the cross sectional area of the nanobridges down to 0.06 $\mu$m$^2$ and investigate the transport properties toward the appearance of the Josephson effects. The critical current $I_c$ measured by the current-voltage characteristics is roughly two orders of magnitude smaller than that for the previous microbridges with larger cross sections [12], while the $I$-$R_n$ product, where $R_n$ is the normal-state resistance along the $c$ axis, is still 3-4 times larger than a theoretical value of Josephson junctions. We also compare between FeTe$_{0.6}$Se$_{0.4}$ and FeSe nanobridges along the $c$ axis, in terms of the occurrence of Josephson effects.

2. Experiments
Fe$_{1-x}$Te$_{0.6}$Se$_{0.4}$ and FeSe single crystals were grown by the self-flux method [15] and the vapor transfer method [16], respectively. The as-grown crystals of Fe$_{1-x}$Te$_{0.6}$Se$_{0.4}$ usually contain some amount (represented by $y$) of excess Fe residing in the interstitial sites of the Te/Se layer. The excess Fe was almost completely removed by the annealing with appropriate amount of Te grains [17]. The superconducting transition temperature $T_c$ of bulk samples is ~14 K for FeTe$_{0.6}$Se$_{0.4}$ and ~9 K for FeSe, respectively.

The $c$-axis nanobridge devices with several cross sectional areas were fabricated by using the FIB techniques, as shown in Figure 1, and are listed in Table 1. In order to largely reduce the cross sectional area of the narrow bridges, in contrast to our previous study [12], a new fabrication method is developed in this work. It is based on the so-called “pickup” method for the preparation of transmission electron microscopy (TEM) samples [18]. First, a small lamella of single crystal is cut into a rectangular shape with a typical size of 10 $\times$ 10 $\mu$m$^2$ and a thickness below 1 $\mu$m, by using the FIB etching. It is picked up and put on a silicon substrate by using a quartz needle. Next, the lamella is wired to Au contact pads by using the Pt deposition induced by FIB. Finally, a narrow bridge part is fabricated between two voltage terminals by using the FIB etching. If desired, the bridge can be locally constricted or weakly divided into two parts to obtain a superconducting weak link, which is necessary for the appearance of Josephson effects. In the previous studies [12,19], the interlayer nanobridge structure of layered superconductors was fabricated between two slits in the sidewall of an in-plane bridge. This suggests that the bridge is perpendicularly standing above the substrate. On the other hand, in the present method, the bridge is always laying on the substrate, which is helpful to avoid a mechanical breakdown of small bridges during the cooling processes to cryogenic temperatures. In addition, both in-plane and interlayer nanobridges can be obtained by choosing a direction of the bridge.

The current-voltage ($I$-$V$) characteristics of the fabricated devices were measured at several temperatures down to 4 K, by applying linearly-ramped bias currents or pulse currents to nanobridges. Other details of the $I$-$V$ measurements are described elsewhere [12].
The fabricated nanobridge devices. Here, $T_c$ is the superconducting transition temperature, which is determined by the zero resistance. $t$ is a lamella thickness, $w$ is a width of the nanobridge, and $S$ is a cross sectional area ($t \times w$). The nanobridges were fabricated by two kinds of trench shape (square or rhombus), as shown in Figure 2 (c).

Table 1. List of the fabricated nanobridge devices. Here, $T_c$ is the superconducting transition temperature, which is determined by the zero resistance. $t$ is a lamella thickness, $w$ is a width of the nanobridge, and $S$ is a cross sectional area ($t \times w$). The nanobridges were fabricated by two kinds of trench shape (square or rhombus), as shown in Figure 2 (c).

| sample name | Composition | $T_c$ (K) | $t$ (µm) | $w$ (µm) | $S$ ($t \times w$) (µm$^2$) | junction shape |
|-------------|-------------|-----------|----------|----------|-----------------------------|----------------|
| int10       | FeTe$_{0.6}$Se$_{0.4}$ | 12.7      | 0.86     | 0.86     | 0.74                        | square         |
| int10s      | FeTe$_{0.6}$Se$_{0.4}$ | 11.9      | 0.86     | 0.32     | 0.28                        | rhombus        |
| int12       | FeTe$_{0.6}$Se$_{0.4}$ | 12.6      | 0.4      | 0.5      | 0.20                        | square         |
| int13       | FeTe$_{0.6}$Se$_{0.4}$ | 10        | 0.35     | 0.17     | 0.06                        | rhombus        |
| int18s      | FeSe        | 8         | 0.4      | 0.69     | 0.28                        | square         |
| int18t      | FeSe        | 7.9       | 0.4      | 0.35     | 0.14                        | square         |
| int18f      | FeSe        | 6.8       | 0.4      | 0.15     | 0.06                        | square         |

3. Results and Discussion

We succeeded in reducing the cross sectional area to 0.06 µm$^2$, which is roughly two orders of magnitude smaller than the values obtained in the previous study [12]. We also note that the widths of the fabricated nanobridges are almost the same as the values for the in-plane nanobridges of FeTe$_{0.5}$Se$_{0.5}$ thin films, where the appearance of Josephson effects were reported [14]. Figures 2(a) and 2(b) show the scanning ion microscopy (SIM) images of the c-axis nanobridge of FeTe$_{0.6}$Se$_{0.4}$ (int13). To realize the superconducting weak links, we tried two kinds of oncoming trench shape, as shown in Figure 2(c). A rhombus shape, which has been used by Wu et al [14], is considered to be more useful to constrict the superconducting wave function as like as a point-contact junction.
Figures 3(a) and 3(d) show the temperature dependences of the c-axis resistance for the smallest nanobridges of FeTe$_{0.6}$Se$_{0.4}$ (imt13) and FeSe (imt18f), respectively. Note that the interlayer resistance of FeCh superconductors is directly measured in the present devices, in contrast to the previous study [12]. The values of $T_c$ for the fabricated nanobridges are slightly smaller than those of bulk samples, suggesting that the superconducting properties in the nanobridge are slightly weakened by the slight inhomogeneity of the residual excess Fe or the small damage due to the FIB etchings. Figures 3(b) and 3(e) show the $I$-$V$ characteristics for imt13 and imt18f, respectively, which were measured at several temperatures from 4 K to $T_c$. For both samples, the $I$-$V$ curves measured at higher temperatures above 5.5 K show the non-hysteresis, similar to the overdamped Josephson junctions, while they change to the finite hysteresis below 5.5 K. Such a hysteretic behavior is due to the Joule heating, as discussed in the previous study [12]. We also find that the critical current $I_c$, determined by the maximum current of the zero-voltage state, is roughly two orders of magnitude smaller than the values obtained for the previous microbridge with the cross section larger than 1 $\mu$m$^2$ [12].

Figures 3(c) and 3(f) show the temperature dependence of $I_c$ and the critical current density $j_c$ for imt13 and imt18f, respectively. The solid lines are the fitting results calculated from Ambegaokar-Baratoff (AB) theory [20],

$$I_c(T) = \frac{\pi \Delta(T)}{2eR_n} \tanh \left( \frac{\Delta(T)}{2k_B T} \right).$$

Here, $R_n$ is the normal-state resistance, $\Delta(T)$ is the superconducting gap, $e$ is the electron charge and $k_B$ is Boltzmann constant. In the plots, we used both of $\Delta(0)$ and $R_n$ as fitting parameters and compared them with the experimental values obtained by the STM studies [21,22] and this study. We find that the fitting values of $\Delta(0)$ are roughly a fifth (imt18f) and half (imt13) of the experimental value and those of $R_n$ is at least one order of magnitude smaller than the experimental values for both samples. Such large differences cannot be resolved by considering the structural difference between the tunnel junctions assumed in the AB theory and the superconducting weak links studied in this work. In other words, the measured $I_c$ is so large that the Josephson junction cannot be formed for both devices. This shows a sharp contrast to the previous study by Wu et al [14]. We also confirm that no step-like structure was observed in the $I$-$V$ curves of both nanobridges, by irradiating several powers of microwaves at 9 GHz and at the lowest temperature.
Figure 3. (Color online) (a) Temperature dependence of the $c$-axis resistance of the FeTe$_{0.6}$Se$_{0.4}$ nanobridge (imt13). (b) The $I$-$V$ curves for imt13, measured at several temperatures below $T_c$. (c) The temperature dependence of $I_c$ (left axis) and $j_c$ (right axis) for imt13. (d) Temperature dependence of the $c$-axis resistance of the FeSe nanobridge (imt18f). (e) The $I$-$V$ curves for imt18f, measured at several temperatures below $T_c$. (f) The temperature dependence of $I_c$ (left axis) and $j_c$ (right axis) for imt18f. Solid orange lines in panels (c) and (f) are the fitting curves obtained by Ambegaokar-Baratoff (AB) theory [20].

Finally, we plot the $I_R$ product of the measured nanobridges as a function of the cross sectional area $S$, as shown in Figure 4. Here, circle (triangle) symbols represent the FeTe$_{0.6}$Se$_{0.4}$ (FeSe) nanobridges. A black (red) horizontal line is a theoretical value for FeTe$_{0.6}$Se$_{0.4}$ (FeSe), which is calculated by using the AB theory [20] and the STM results [21,22]. The calculated values of $I_R$ are 2.6 mV for FeTe$_{0.6}$Se$_{0.4}$ and 3.6 mV for FeSe, respectively. The results indicate that the obtained $I_R$ products steadily approach to the theoretical values of Josephson junctions with decreasing $S$. Particularly, the $I_R$ product of the FeTe$_{0.6}$Se$_{0.4}$ nanobridge is largely decreased below $S = 0.1 \, \mu m^2$. This suggests that the measured critical current begins to deviate from the depairing current regime and approaches to the Josephson current regime, expecting that the further reduction of $I_R$ product will trigger the appearance of Josephson effects. On the other hand, we find that the decreasing rate for the FeSe nanobridges is smaller than that for FeTe$_{0.6}$Se$_{0.4}$ nanobridges. The magnitude of the $I_R$ product of the FeSe nanobridges with $S > 0.1 \, \mu m^2$ is roughly one order of magnitude smaller than that of the FeTe$_{0.6}$Se$_{0.4}$ nanobridges. This seems to be consistent with the stronger two-dimensionality in the electronic state of FeSe than that of FeTe$_{0.5}$Se$_{0.5}$, suggested by the first-principles calculations based on the density functional theory [23], where the electronic band dispersion along the $\Gamma$-$Z$ direction of FeSe is much weaker than that of FeTe$_{0.5}$Se$_{0.5}$. However, below $S = 0.1 \, \mu m^2$, the decrease of the $I_R$ product of the FeSe nanobridge is rather small, in contrast to the case of the FeTe$_{0.6}$Se$_{0.4}$ nanobridge. This reason is unknown at the present. Nevertheless, the $I_R$ products for the smallest nanobridges (imt13, imt18f) are still three or four times larger than the theoretical limits to the Josephson current regime. Actually, the critical current density for the FeTe$_{0.6}$Se$_{0.4}$ nanobridges (imt13) is still large ($1.3 \times 10^5 \, A/cm^2$ at $T = 4 \, K$), since it is roughly a half of the $c$-axis depairing current density determined by the previous study [12]. Therefore, further reduction
of the $I_cR_n$ products is required to realize the single-crystal Josephson device made of FeCh superconductors.

![Figure 4](image)

**Figure 4.** (Color online) The plots of the $I_cR_n$ products as a function of the cross sectional area of the measured nanobridges. Solid circles (triangles) are FeTe$_{0.6}$Se$_{0.4}$ (FeSe) samples. Horizontal black (red) lines are the theoretical values for FeTe$_{0.6}$Se$_{0.4}$ (FeSe), calculated by using the Ambegaokar-Baratoff theory [20] and the STM results [21,22].

**4. Conclusion**

We present a study on the transport properties of the c-axis nanobridges with several cross sectional areas, which were fabricated from FeTe$_{0.6}$Se$_{0.4}$ and FeSe single crystals by using the FIB techniques. In order to largely reduce the cross sectional area of the narrow bridges, the fabrication method was newly developed in this work. Unfortunately, the reduction of the cross sectional area down to 0.06 $\mu$m$^2$ is still insufficient for the appearance of the Josephson effects in the c-axis nanobridge devices of FeTe$_{1-x}$Se$_x$ single crystals. This shows a sharp contrast to the previous report [14] on the in-plane nanobridges made of FeTe$_{0.5}$Se$_{0.5}$ thin films, suggesting the difficulty of the single-crystal Josephson devices. We focus on the dependence of the $I_cR_n$ products for the measured nanobridges on the cross sectional area $S$, indicating the decrease of the $I_cR_n$ products with decreasing $S$. Probably, the further reduction of the $I_cR_n$ product will lead to the realization of single-crystal Josephson devices of FeCh superconductors. Very recently, we carefully made a short cut on the FeTe$_{0.6}$Se$_{0.4}$ nanobridge (a width is 0.12 $\mu$m, a thickness is 0.24 $\mu$m) and filled the Pt deposit in the gap (roughly 30 nm), forming a superconductor/normal metal/superconductor (SNS) structure. Preliminary results show no sign of Josephson effects, since the gap is too large and the amount of Pt deposit is too much. This suggests that more precise control of the FIB etching and deposition is needed to realize SNS-type Josephson junctions.

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References

[1] Hsu F C, Luo J Y, Yeh K W, Chen T K, Huang T W, Wu P M, Lee Y C, Huang Y L, Chu Y Y, Yan D C and Wu M K 2008 Proc. Nat. Acad. Sci. 105 14262

[2] Sales B C, Sefat A S, McGuire M A, Jin R Y, Mandrus D and Mozharivskyi Y 2009 Phys. Rev. B 79 094521

[3] Medvedev S, McQueen T M, Troyan I A, Palasyuk T, Eremets M I, Cava R J, Naghavi S, Casper F, Ksenofontov V, Wortmann G and Felser C 2009 Nat. Mater. 8 630

[4] Burrard-Lucas M, Free D G, Sedlmaier S J, Wright, J D, Cassidy S J, Hara Y, Corkett A J, Lancaster T, Baker P J, Blundell S J and Clarke S J 2013 Nat. Mater. 12 15

[5] He S, He J, Zhang W, Zhao L, Liu D, Liu X, Mou D, Ou Y B, Wang Q Y, Li Z, Wang L, Peng Y, Liu Y, Chen C, Yu L, Li G, Dong X, Zhang J, Chen C, Xu Z, Chen X, Ma X, Xue Q and Zhou X J 2013 Nat. Mater. 12 605

[6] Lubashevsky Y, Lahoud E, Chashka K, Podolsky D and Kanigel A 2012 Nat. Phys. 8 309

[7] Okazaki K, Ito Y, Ota Y, Kotani Y, Shimojima T, Kiss T, Watanabe S, Chen C T, Niitaka S, Hanaguri T, Takagi H, Chainani A and Shin S 2014 Sci. Rep. 4 4109

[8] Zhang P, Yaji K, Hashimoto T, Ota K, Kondo T, Okazaki K, Wang Z, Wen J, Gu G D, Ding H and Shin S 2018 Science 360 182

[9] Zhang P, Wang Z, Wu X, Yaji K, Ishida Y, Kohama Y, Dai G, Sun Y, Bareille C, Kuroda K, Kondo T, Okazaki K, Kindo K, Wang X, Jin C, Hu J, Thomale R, Sumida K, Wu S, Miyamoto K, Okuda T, Ding H, Gu G D, Tamegai T, Kawakami T, Sato M and Shin S 2019 Nat. Phys. 15 41

[10] Wang D, Kong L, Fan P, Chen H, Zhu S, Liu W, Cao L, Sun Y, Du S, Schneeloch J, Zhong R, Gu G, Fu L, Ding H and Gao H J 2018 Science 362 333

[11] Machida T, Sun Y, Pyon S, Takeda S, Kohsaka Y, Hanaguri T, Sasagawa T and Tamegai T 2019 Nat. Mater. 18 811

[12] Sun Y, Ohnuma H, Ayukawa S, Noji T, Koike Y, Tamegai T and Kitano H 2020 Phys. Rev. B 101 134516

[13] Moll P J W, Zhu X, Cheng P, Wen H-H and Batlogg B 2014 Nat. Phys. 10 644

[14] Wu C H, Chang W C, Jeng J T, Wang M J, Li Y S, Chang H H, and Wu M K 2013 Appl. Phys. Lett. 102, 222602

[15] Sun Y, Yamada T, Pyon S and Tamegai T 2016 Sci. Rep. 6 32290

[16] Sun Y, Pyon S and Tamegai T 2016 Phys. Rev. B 93 104502

[17] Sun Y, Shi Z, Tamegai T 2019 Supercond. Sci. Technol. 32 103001

[18] Moll P J W 2018 Ann. Rev. Condens. Matter Phys. 9 147

[19] Kakizaki Y, Koyama J, Yamaguchi A, Umemag S, Ayukawa S and Kitano H 2017 Jpn. J. Appl. Phys. 56 043101

[20] Ambegaokar V and Baratoff A 1963 Phys. Rev. Lett. 10 486

[21] Hanaguri T, Niitaka S, Kuroki K and Takagi H 2010 Science 328 5977

[22] Kasahara S, Watashige T, Hanaguri T, Kohsaka Y, Yamashita Y, Shimoyama Y, Mizukuri Y, Endo R, Ikeda H, Aoyama K, Terashima T, Uji S, Wolf T, Löhneysen V H, Shibauchi T and Matsuda Y 2014 Proc. Natl. Acad. Sci. USA 111 16309

[23] Wang Z, Zhang P, Xu G, Zeng L K, Miao H, Xu X, Qian T, Weng H, Richard P, Fedorov A V, Ding H, Dai X, and Fang Z 2015 Phys. Rev. B 92 115119