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superconducting FeSe_{0.5}Te_{0.5} thin films by
low temperature oxygen annealing**

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2 **Doubling the Critical Current Density in Superconducting FeSe_{0.5}Te_{0.5}**

3 **Thin Films by Low Temperature Oxygen Annealing**

4

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11

12 **Abstract**

13 Iron-chalcogenide superconducting thin films and coated conductors are attractive for potential
14 high field applications at liquid helium temperature for their high critical current densities J_c ,
15 low anisotropies, and relatively strong grain couplings. Embedding flux pinning defects is a
16 general approach to increase the in-field performance of superconductors. However, many
17 effective pinning defects can adversely affect the zero field or self-field J_c , particularly in
18 cuprate high temperature superconductors. Here, we report doubling of the self-field J_c in
19 FeSe_{0.5}Te_{0.5} films by low temperature oxygen annealing, reaching ~ 3 MA/cm². In-field
20 performance is also dramatically enhanced. Our results demonstrate that low temperature
21 oxygen annealing is a simple and cost-efficient post-treatment technique which can greatly help
22 to accelerate the potential high field applications of the iron-based superconductors.

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1 Iron-based superconductors have attracted great interests since their discovery in 2006 by
2 Kamihara *et al.*¹ They are superconducting materials with transition temperature, T_c , up to 100 K in
3 monolayer thin films, and very high upper critical field above 100 T.²⁻⁸ Combined with low
4 anisotropies in magnetic field, this class of superconductors is considered very promising for high
5 field applications. A key limiting factor for the iron based superconductors is the critical current
6 density, J_c , that still needs to be raised at or above the level of the competing superconductors.
7 Currently, cuprate high- T_c superconductors (HTS) are the dominating one in terms of the J_c
8 performance.⁷ However, the cuprate HTS have intrinsic high anisotropy and low tolerance of grain
9 boundary misalignment. When these drawbacks coupled with high processing cost, cuprate HTS
10 become less competitive than the low temperature Nb₃Sn in high-magnetic field (up to 14 T)
11 applications at liquid helium temperature on the cost-performance basis. The recent developments of
12 thin film iron-based superconductors and coated conductors have provided an alternative
13 superconducting material potentially being used in the next generation high field devices. High
14 self-field J_c over 4MA/cm² at 4.2K has been observed in iron pnictide “122” thin films.⁹⁻¹¹ High
15 self-field J_c above 1MA/cm² was also reported in iron chalcogenide films.^{12,13} Most recently, a high
16 field isotropic J_c has been observed in FeSe_{0.5}Te_{0.5} thin films grown on CaF₂ substrates with a
17 self-field J_c over 2MA/cm² at 5K.¹⁴

18 There are some concerns for the practical application of the iron-based superconductors. For
19 example, the toxic element As in the most studied system, iron pnictides, is one of them, although the
20 superconducting iron pnictides have a relatively higher T_c and J_c among iron-based superconductors.
21 The remaining magnetic excess Fe in the synthesis of the iron based superconductors is another issue,
22 as they can act as the Cooper pair breakers that severely weaken superconductivity.^{15,16} Report has
23 shown that 3% of excess Fe will totally suppress superconductivity in FeSe, which normally has a T_c

1 $\sim 8\text{ K}$ ¹⁷. The sensitivity of the superconducting properties to the excess Fe makes it difficult to
2 produce the reliable iron chalcogenide samples of consistent quality.

3 Efforts have been made by post-treatment to remove the excess Fe. It has been reported that
4 the excess Fe can be effectively reduced through reaction with acid, oxygen, even alcohol
5 beverages¹⁸⁻²⁶. Among these methods, oxygen annealing is found to be most effective.
6 Enhancements on both T_c and J_c were observed in annealed $\text{FeSe}_{1-x}\text{Te}_x$ single crystals.^{21,25,26} Yet the
7 study of the annealing effect on thin films of iron-based superconductors, which mostly have higher T_c
8 and J_c than those of bulk, is still lacking.

9 Here we report a strong enhancement of superconducting properties in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ (FST) thin
10 films, through low temperature oxygen annealing. Iron chalcogenides, known as the “11” system, are
11 an interesting sub-class of iron-based superconductors. They have the simplest crystal structure. J_c
12 anisotropy of the $\text{FeSe}_{1-x}\text{Te}_x$ films in magnetic field can be as low as 2, lower than that of pnictides⁷.
13 The optimum superconductivity was found in $\text{FeSe}_{1-x}\text{Te}_x$ when $x \cong 0.5$, with a $T_c = 14.5\text{ K}$ in bulk
14 materials. Our previous study shows an enhanced T_c and J_c in FST thin films made by pulsed laser
15 deposition (PLD)^{12,17}. With a CeO_2 buffer, the films exhibit a self-field J_c above 1 MA/cm^2 at 4.2 K . In
16 this letter, we show that low temperature oxygen annealing can enhance J_c dramatically. At 5 K ,
17 self-field J_c in the annealed films can reach 3 MA/cm^2 .

18 The FST thin films were grown on single crystal substrates by PLD with a CeO_2 buffer. The
19 films are of 3 mm by 5 mm and the thickness of the films is around 130 nm . The detailed film synthesis
20 has been reported previously¹². We performed oxygen and vacuum annealing on two sets of films.
21 The oxygen annealing was carried out sequentially at 90°C in 100 mTorr oxygen environment in four
22 steps with the annealing time of 15, 30, 60 and 120 minutes. We found that higher oxygen annealing
23 temperature than 90°C weakens superconductivity in these films. Vacuum annealing at $1 \times 10^{-8}\text{ Torr}$

was done at 200°C in three steps with the annealing time of 15, 30 and 60 minutes. Non-destructive magnetization measurement, with magnetic field perpendicular to the film surface, was performed to characterize the samples after growth and each step of the annealing to compare the changes of superconducting properties. Magnetization measurement is particularly suitable for continuously monitoring the property change. The derived J_c values from the magnetic hysteresis measurement based on the Bean critical state model²⁸ were compared with the J_c values obtained directly from the transport measurement on the laser patterned film bridges, and their differences are within 10%.

Fig. 1(a) shows the magnetic field dependence of J_c at 5 K for one of the sequentially annealed FST films in oxygen for different annealing time, where magnetic field is applied along the c-axis of the film. Inset shows the magnetization measurement of the films under zero field cool (ZFC) and field cool (FC) for $B = 2$ gauss ($//$ c-axis). It is apparent that a short term oxygen annealing remarkably enhances J_c in all fields. The J_c values can be further enhanced by increasing the annealing time before saturated at the last step of two hours annealing – upon which oxygen annealing for longer time does not further enhance the J_c . Fully oxygen annealing the film had the self-field J_c value more than doubled, from 1.24MA/cm² in the as grown films to 2.54MA/cm², as shown in Fig. 1(a). In-field J_c enhancement is much more impressive - more than 300% increase is achieved at fields above 1.5T. Fig. 1(b) shows the magnetic field dependence of the J_c enhancement,

defined as $\frac{J_c^{\text{annealed}} - J_c^{\text{as-grown}}}{J_c^{\text{as-grown}}}$. The process has been repeated in several films, where a similar

magnitude of J_c enhancement was observed, with the highest self-field J_c reaching 3MA/cm² at 5K. While J_c is enhanced by oxygen annealing, we notice a small and gradual reduction of T_c as the annealing time increases. This is shown in the inset of Fig. 1(a). After two hours oxygen annealing, the onset T_c reduces from 17.6K to 16.2K, but the transitions becomes sharper.

Fig. 2 shows the magnetic field dependence of J_c and the J_c enhancement at 5K for one of

1 the sequentially annealed FST films in vacuum for different annealing time. The change of T_c of the
2 film after vacuum annealing, shown in the inset, similar to that of oxygen annealed sample. We found
3 the magnitude of J_c enhancement by vacuum annealing saturates at annealing time of ~ 1 -2 hours, and
4 is much less than that by the oxygen annealing. The self-field J_c of this fully vacuum annealed film is
5 $1.38\text{MA}/\text{cm}^2$, $\sim 38\%$ above that of the as grown films, and in-field J_c enhancement is up to 180%.

6 It is evident that the annealing environment plays a significant role in the film's J_c
7 performance. To further demonstrate that oxygen annealing is superior, we gave the last vacuum
8 annealed film (solid triangle in Fig. 2) an additional 2-hour oxygen annealing at 90°C . The result
9 (open circle) is plotted in Fig. 2 for comparison. Additional oxygen annealing leads to the self-field J_c
10 being greatly improved to $2.27\text{MA}/\text{cm}^2$ and again about 300% enhancement in the in-field J_c on
11 average was observed, which is similar to that of the fully oxygen annealed film (open circles) as seen
12 in Fig. 1. We notice little or no change in T_c after additional oxygen annealing.

13 Let us discuss the possible origin of the observed J_c enhancement. One of the explanations
14 could be the removal of the excess Fe in the FST films. It is known that in bulk FST, excess Fe atoms
15 are magnetic, and act as the Cooper breakers and suppress superconductivity.^{21,25} In thin films,
16 pair-breaking effect by interstitial Fe is suspected to be much stronger due to its low dimension. It is
17 postulated that the excess Fe in our films, although very small, probably reacts with oxygen to form
18 non-magnetic oxides during the sequential annealing, in a similar way to that found in another
19 report²⁵. The increase of J_c may also due to the phase change induced by interstitial oxygen. Similar
20 oxygen annealing studies in $\text{Fe}_{1.08}\text{Se}_{0.45}\text{Te}_{0.55}$ single crystals²⁹ and Fe_{1+y}Te thin films³⁰ have been
21 reported, where improved superconductivity emerges with the formation of the new phase after
22 oxygen annealing. It is also possible that both mechanisms are effective at the same time. We have
23 investigated the annealed films extensively through structural characterizations. Our TEM

examination did not shown any iron oxides. No observable phase change, like Hu *et al* had seen in $\text{Fe}_{1.08}\text{Se}_{0.45}\text{Te}_{0.55}$ single crystals, can be observed in our films either. It could be due to the very small amount of the excess Fe in our films, since our films were grown from stoichiometric $\text{FeSe}_{0.45}\text{Te}_{0.55}$ targets without intentionally introducing excess Fe.

We note that the T_c in our FST films was slightly suppressed after oxygen annealing. This result is different from what happened in oxygen annealed single crystals, where an enhanced T_c was reported.^{19,23-25} The suppression of T_c in our FST films is found in both oxygen and vacuum annealing process. One possible explanation is that the fast cooling after the film growth causes the lattice inhomogeneity in the film and creates imbalanced local strain. It has been reported by Bellingeri *et al*^{19, 31,32} that the compressive strain in *ab* plane can enhance the T_c of FST films. In this scenario, the inhomogeneity in the film may lead to a distribution of the strain field, and hence T_c . The film may comprise of higher T_c regions with compressive strain and lower T_c regions with tensile strain. The inhomogeneity in the as-grown films results in a relatively wider superconducting transition as shown in the insets of Fig. 1(a) and Fig. 2(a). One of the consequences of the sequential annealing, either in oxygen or vacuum, is the release of strain in the FST films, leading to more homogenous strain distribution. This is consistent with the sharper superconducting transition and slightly lowered on-set T_c observed in the annealed samples.

Considering the fact that the J_c of the films increases significantly at an annealing temperature as low as 90°C with the presence of oxygen, one could expect a qualitatively similar J_c enhancement by aging in the air at room temperature. Oxygen in the air should slowly react with the freshly grown films. This was indeed the case when we measured the J_c of the fully oxygen-annealed films and the as-grown films after one year stored in a dry environment, but in contact with the air at room temperature. The results of the field dependent J_c before and after one year aging are shown in Fig. 3a.

1 For the as-grown film, the self-field J_c increases from $0.96\text{MA}/\text{cm}^2$ to $1.53\text{MA}/\text{cm}^2$ and the in-field J_c
2 increases by 90% ~ 100% from 2T to 4.5T, in qualitative agreement with the oxygen annealing. For
3 the fully oxygen annealed film, the one-year aging produce virtually no effect - the field dependent J_c
4 difference is less than 5%. This observation is in line with both explanations mentioned previously.
5 The oxygen in the air can react with the as-grown film, leading to excess Fe removal or the phase
6 change, but have no effect on fully oxygen annealed films.

7 For a comprehensive understanding, we also conduct a 2-hour vacuum annealing on a fully
8 oxygen annealed film in order to check the irreversibility of the oxygen annealing effect. The result
9 is shown in Fig. 3b, where black squares, red triangles and blue open circles stand for the field
10 dependent J_c of the film before annealing, after fully oxygen annealing and after an additional
11 vacuum annealing, respectively. It is seen that the vacuum annealing shows virtually no influence
12 on J_c after the film was fully oxygen annealed. The J_c enhancement caused by the oxygen is
13 irreversible.

14 Fig. 4 shows the direct transport J_c of an oxygen annealed FST film as a function of
15 magnetic field at different temperatures, together with the magnetization-derived J_c at $T = 5\text{K}$ (solid
16 circles in Fig. 3). The transport measurement was taken after the magnetization measurement by
17 patterning the same film into micro bridges, $300\mu\text{m}$ long and $10\mu\text{m}$ wide. J_c was obtained from I-V
18 curves by using $1\mu\text{V}$ criteria, and a typical one measured at 5K under 0.1T field is shown in the
19 lower inset of Fig.4. We note the J_c values obtained by the transport measurement and
20 magnetization measurement are virtually the same. Upper inset shows the superconducting
21 transition at 0, 1, 3, 5, 9 Tesla by the resistance measurement. The zero resistance T_c is 17K at zero
22 field and 14.5K at 9T, applied perpendicular to the film surface.

23 Figs. 5(a) and 5(b) present the data of the in-field ($B//c$) J_c and the pinning force, $F_p (= J \times B)$

1 in fully oxygen annealed films at $T = 5$ K, together with the FST coated conductor grown on the
 2 rolling-assisted biaxially textured substrate (RABiTS)¹² and several prominent superconducting
 3 materials (4.2 K data), including the second-generation (2G) $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) wire^{33,34},
 4 thermo-mechanically processed Nb47Ti^{35,36} and small grain Nb_3Sn wire^{37,38}. The solid lines in Fig.
 5 5(b) are plotted using Kramer's scaling approximations³⁹. In general, J_c and F_p of the FST films are
 6 higher than that of the Nb-Ti wires, but substantially lower than the 2G YBCO coated conductors. It is
 7 interesting to compare the present results with the FST coated conductors. The oxygen annealed films
 8 have higher J_c and F_p at low fields, but lower J_c and F_p at high fields. Although we do not fully
 9 understand the pinning mechanism in the FST coated conductors at present, a higher performance
 10 FST coated conductor may be anticipated when its strong pinning at high field is combined with the
 11 enhanced pinning achieved at low field by the practical and cost-efficient oxygen annealing, which
 12 doubles the self-field J_c . The dotted line in Fig. 5 shows an estimated J_c value we can expect by
 13 applying a lifting factor, derived from the magnitude of the J_c enhancement obtained through the
 14 oxygen annealing, to the J_c value known for the as-grown FST coated conductor on RABiTS. The
 15 proposed optimally annealed FST coated conductor would have a comparable performance with the
 16 Nb_3Sn below 4T, and much better above, and that would represent a significant step forward in the
 17 potential high field applications of the iron based superconductors at liquid helium temperature.

18 In summary, short time (~ 1 -2 hours) oxygen annealing at low temperature (below 100°C)
 19 has been found to be very effective on the J_c enhancement in the FST superconducting thin films.
 20 The fully annealed FST thin films exhibit a doubled J_c at self-field and four-fold J_c enhancement
 21 above 1.5T. Possible explanations for the observed J_c enhancement in FST films could be the reaction
 22 between the excess Fe and oxygen, or the phase change due to the interstitial oxygen – both were
 23 previously found effective to improve the superconductivity in the form of single crystals.

1 Qualitatively, similar J_c enhancement is also achieved by room temperature aging in the air, which is
2 likely due to the same underline mechanism with the oxygen annealing. The reaction between the
3 FST film and oxygen is found irreversible. The oxygen annealing process identified in this study can
4 serve as a fast, easy, and low-cost approach to effectively enhance the J_c in the iron based
5 superconductors. Using this process, the highest self-field J_c of $3\text{MA}/\text{cm}^2$ at 5K was achieved in the
6 iron chalcogenides. Combined with other growth induced pinning defects, or artificial pinning
7 centers, such as these produced by the proton or ion irradiation, iron chalcogenide coated conductors
8 are very attractive for the high field application at liquid helium temperature.

9
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Figures and Legends

Figure 1

Figure 1: Magnetic field dependence of J_c (a) and J_c enhancement (b) of one of the sequentially annealed FST films in 100mTorr oxygen environment for different annealing time. Inset in (a) shows the T_c change by the magnetization measurement of the film under zero field cool (ZFC) and field cool (FC). Inset in (b) shows a typical half magnetization loop from which the field dependent magnetization J_c is derived.

Figure 2

Figure 2: Magnetic field dependence of J_c (a) and J_c enhancement (b) of one of the sequentially vacuum annealed FST films. Inset shows the T_c changes by the magnetization measurement of the film under zero field cool (ZFC) and field cool (FC). The result of the additional two-hour oxygen annealing at 90°C following the vacuum annealing is also plotted for comparison (open circles).

Figure 3

Figure 3: (a) Magnetic field dependence of J_c of the FST films upon aging for one year. J_c increases in the as-grown film, while no change in the fully oxygen annealed film. (b) Magnetic field dependent J_c of a FST film which was first fully oxygen annealed and then vacuum annealed. The additional vacuum annealing have virtually no further influence on J_c , indicating that the J_c enhancing process caused by oxygen is irreversible.

1 **Figure 4**

2
3 Figure 4: Magnetic field dependence of J_c of a fully oxygen annealed FST film at different
4 temperatures by transport measurement. J_c (solid circles) obtained from magnetic hysteresis
5 measurement of the same film at $T = 5\text{ K}$ is also plotted for comparison. Upper inset shows the
6 superconducting transition at various fields (0, 1, 3, 5, 9 Tesla) by the resistive measurement. Lower
7 inset shows a typical I-V curve from which the critical current density was extracted using $1\mu\text{V}$
8 criteria.

1 **Figure 5**

2
3 Figure 5: Magnetic field dependence of J_c (a) and vortex pinning force F_p (b) of several high field
4 superconductors, including oxygen annealed FST film (this work), the FST coated conductors on
5 RABiTS (labeled as “Ra”)¹², Nb-Ti, Nb₃Sn, and 2G YBCO. The lines in (b) are fitting curves using
6 the Kramer’s scaling approximation. 4.2K J_c data of YBCO, Nb₃Sn and Nb-Ti are adopted from
7 literatures³³⁻³⁸. The red dotted line shows a practical target to achieve in the near future for the FST
8 coated conductors.