nature portfolio

Peer Review File

Oriented Bi2Te3-based films enabled high performance planar thermoelectric cooling device for hot spot elimination

Corresponding Author: Professor Ruiheng Liu

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
“High Orientated Bi2Te3-based Film Leads to High-Performance Planar Thermoelectric Cooling Device for Hot Spot Elimination” by Dong et al.

Overall merits

This manuscript presents some significant advances in the field of thermoelectric materials. The introduction of a novel Te-assisted growth method to achieve highly oriented Bi2Te3-based films is a notable innovation, contributing to the ongoing efforts to enhance the efficiency and applicability of thermoelectric devices.

The reported in-plane zT values of ~1.53 for P-type Bi0.4Sb1.6Te3 and ~1.10 for N-type Bi2Te3 films are particularly impressive and represent a substantial improvement in thermoelectric performance, approaching those of bulk materials and single-crystal nanosheets.

The application of these films in planar thermoelectric cooling devices (f-TECs) is also a noteworthy contribution, with a significant temperature reduction of ~8.2 K in hot spot elimination experiments. This work not only shows potential for practical applications in electronics cooling but also contributes valuable insights into the design and synthesis of high-performance thermoelectric materials.

I would recommend the manuscript be accepted subject to a major revision, addressing the following questions and suggested improvements.

Questions and specific suggested improvements

(1) Although the Te-assisted growth method is innovative, further optimization could be explored, for example, what effects of annealing temperatures and Te concentrations will have on the orientation and zT values.

(2) What are the long-term stability and repeatability of the film growth process?

(3) The microstructural analysis, while thorough, could be enhanced by a deeper exploration of how the observed microstructural features specifically influence phonon scattering and thermal conductivity. Correlating these structural properties with mechanical stability under operational stress conditions would also offer valuable insights.

Reviewer #2

(Remarks to the Author)
How to improve the texture degree of the (00l) plane is a very important issue for Bi2Te3-based thermoelectric films. This work intelligently proposed an extra-Te-assisted method to achieve high (00l) orientation of both p- and n-type Bi2Te3-based films and profoundly discovered the mechanics of grain growth with extra Te by the chemical bonding theory. This method produces the orientation of polycrystal film to be comparable with those of single sheets, which gets effect instantly

in the improvement of the electrical properties. On the other hand, the extra Te in the film provided by this method induces rich microstructures, such as dislocations and stacking faults, which can strongly depress the thermal conductivity. Finally, exciting performances of both Bi2Te3-based films (zT=1.5) and the film device (ΔT=~8.2 K) have been achieved. Both the result and analysis are solid and organized well. Before the manuscript is accepted, there are still some questions that need to be solved.
(1) The high texture degree of the (00l) plane produces high carrier mobility by this extra-Te-assisted method, but this advantage has been not strongly illustrated for the lack of necessary comparison.
(2) The extra Te can induce the Te–(BiSb)–Te layers and Te–(BiSb)–Te–(BiSb)–Te–(BiSb)–Te seven-layers, but it little affects the carrier mobility, can the author provide a relevant explanation? Besides, is there evidence to verify the similar compositions of x=0.17 and x=0.24 after annealing?

(3) The words in Fig.1a are in disorder, please rearrange the words properly.
(4)“... that the growth rate along (00l) planes is almost ten times higher than that of vertical (00l) planes, and also about 3 times higher than that along (015) planes, as shown in Figure 2...” the expression of (00l) planes and (015) planes should be corrected.
(5) The EDS in Fig. S7 shows Te second phase between grain boundaries, however, the authors claimed extra Te has evaporated after annealing at the last of page 4, please explain the inconsistence.
(6) The format of the figure 6 should be consistent with other figures.

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In this work, highly (00l)-oriented Bi2Te3 films, comparable to single crystals, were obtained using a liquid Te-assisted growth method. The high mobility resulting from the orientation, combined with low lattice thermal conductivity due to Te-induced staggered stacking faults, led to in-plane zT values of ~1.53 for P-type Bi0.4Sb1.6Te3 and ~1.10 for N-type Bi2Te3 films. Planar f-TEC devices based on these films achieved a temperature reduction of ~8.2 K in hot spot elimination experiments, demonstrating the potential of this method for producing oriented planar Bi2Te3 films and for f-TEC device design, with important implications for next-generation thermal management in integrated circuits. Therefore, I recommend its publication in Nature Communication after addressing the following:
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2. Some typos：line 51 ， 2, 5；line 66 powder factor；line 94 ，comparasion ，leaser ，etc
3. Please carefully check if the data mentioned in the text matches the figures. For example, verify whether the value of 0.08 in "F jumps from 0.08 for x = 0 to 0.84 for x = 0.24" on lines 127-128 corresponds correctly with the data shown in the figures.
4. Please provide the manufacture information of Magnetron sputtering device. And how long it takes to deposit the BT films?
5. What is the reason for the gap between the experiment and the simulated values in Figure 6? How long could the high-record delta T keep when the laser and TEC is on together?

Version 1:

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The authors have satisfactorily addressed the issues and concerns I raised in my initial review. I think the paper can be published now

Reviewer #2

(Remarks to the Author)
I believe the author has addressed my concerns, and the article is now logically sound and acceptable.

Reviewer #3

(Remarks to the Author)
Publish as is

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I would recommend the manuscript be accepted subject to a major revision, addressing the following questions and suggested improvements.

Response: Thanks for the recommendation.

Questions and specific suggested improvements

(1) Although the Te-assisted growth method is innovative, further optimization could be explored, for example, what effects of annealing temperatures and Te concentrations will have on the orientation and zT values.

Response: Thanks for your suggestions. The effect of Te-assistant growth method is to provide extra Te for in-plane crystal growth and then improve in-planed orientation. The samples must be annealed at the temperature near Te melting point (720 K), which can provides the liquid-sintering effect to accelerate the in-plane growth speed of grains. Indeed, we have further carried out the experiment that annealing the as-deposited $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.24) films and $Bi_2Se_{0.3}Te_{2.7+x}$ (x=0.12) at different temperatures as displayed Fig. R1 below. It is shown that the orientation greatly enhanced when annealing temperature is above 673 K. Therefore, elevating the annealing time greatly helps to enhance the mobility and the final zTs.

The XRD and orientation results of $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.24) and $Bi_2Se_{0.3}Te_{2.7+x}$

(x=0.12) sample annealing at different temperature are added in the Supporting Information Fig.S5.

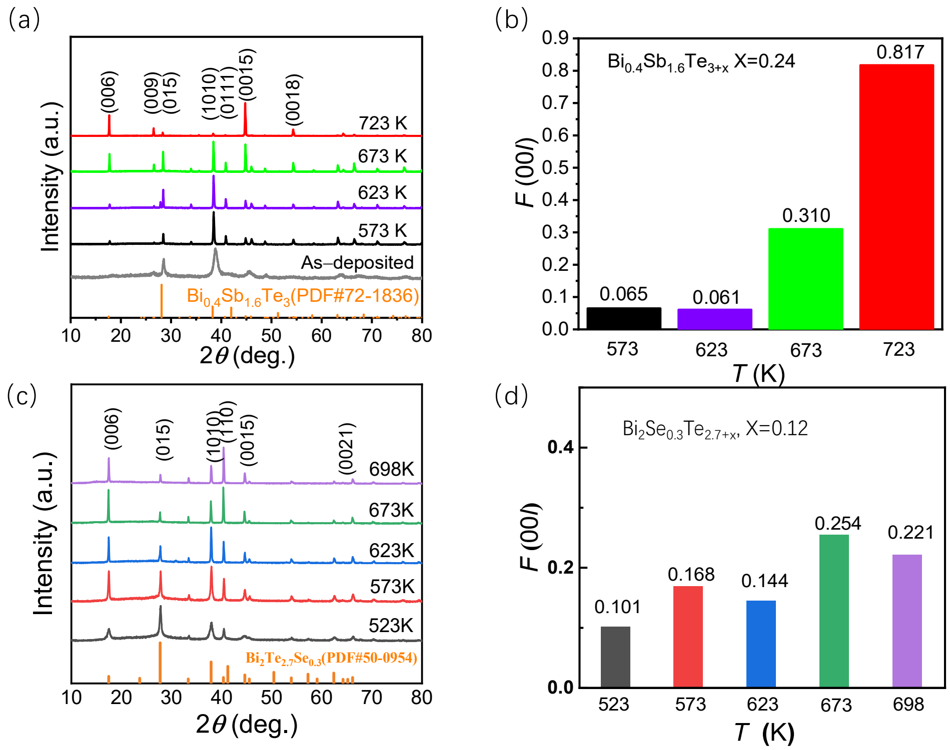


Fig. R1. The XRD patterns and (00*l*) orientation factor (b, d) of $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.24), and $Bi_2Se_{0.3}Te_{2.7+x}$ (x=(a, c)0.12) samples annealed at different temperature, respectively.

Meanwhile, Te content would influence the carrier concentration for both P-type and N-type films, and high annealing temperature would also greatly affects defects state, and further determines the carrier concentration. We have investigated the effect of Te content from x=0 to x=0.24 on the carrier concentration, as shown by the updated Fig. S14 in Supporting Information. For P-type $Bi_{0.4}Sb_{1.6}Te_3$ films, more Te content would slightly increase the hole concentration since Te acts as acceptor, and the concentration would be stably approach 2.0E19 cm^{-3} as Te content tend to saturation. However, for N-type films, if the Te were not supplemented (x=0), the lack of Te induced by evaporation would enhance the electron concentration, and with the increase of Te content, the carrier concentration firstly decrease, but it will then increase if further supplementing Te, since more Te content would increase the density of Bi-Te antisite defects which act as donor in n-type Bi_2Te_3 materials, making n-type films more hardly to optimized. We still working on the annealing process on the carrier concentration adjustment for ternary N-type $Bi_2Te_3Se_{3-x}$ films with extra Te right now, and will discuss this questions in the future.

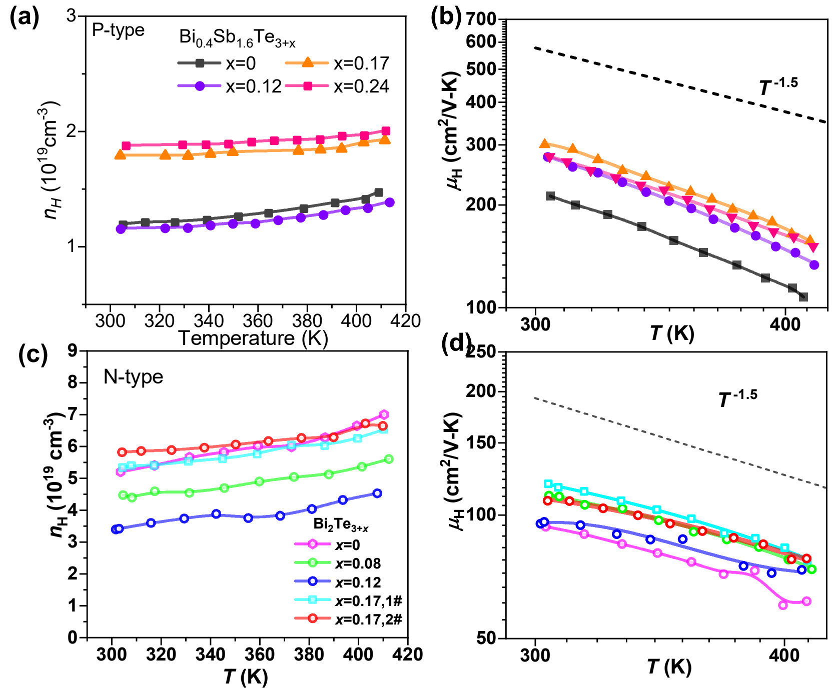


Fig. R2&S14. **Electrical properties for p- and n-type Bi_2Te_3-based samples.** The temperature dependent hall carrier concentration and hall mobility for *p*-type $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0, 0.12, 0.17 and 0.24) (a~b) and *n*-type Bi_2Te_{3+x} (x=0, 0.08, 0.12 and 0.17) (c~d) films in this work.

(2) What are the long-term stability and repeatability of the film growth process?

Response: Thanks for the concern.

In this study, the films were obtained by Magnetron co-sputtering and post-annealing process. Firstly, two targets of $Bi_{0.4}Sb_{1.6}Te_3$ target and Te target were adopted for P-type $Bi_{0.4}Sb_{1.6}Te_3$ films, and Bi_2Te_3 target and Te target for N-type Bi_2Te_3 films. The total deposition time need to reach 135 mins for the 4 μm thick $Bi_{0.4}Sb_{1.6}Te_3$ and Bi_2Te_3 films. We also paid much attention to the sputtering stability since with the thickness increases above 1.0 um, the composition maybe deviate the stoichiometric ratio of compound targets. We have studied the sputtering time dependent compositions of the as-deposited films before and after annealing, as shown in the following Table R1. At the beginning of sputtering (within 30~40mins), the Te content is relatively lower, and it would increase with prolonging the sputtering time and tend to stable. Meanwhile, after annealing, all the samples composition were very close to the stoichiometric ratio since the most of extra Te were evaporated, which leads to rather good composition repeatability of final samples.

Table R1. the EDS results of Bi_2Te_{3+x} (x=0.17) and $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.24) before and after sputtering process, respectively.

Samples	Supply power for compounds	Supply power for Te	Sputtering time (min)	Thickness (μm)	Te content before	Te content after

	target	target (W)			annealing (at%)	annealing (at%)
Bi_2Te_{3+x} x=0.17	90	30	45	1.5	61.54	59.50
			100	3.4	62.50	60.50
			135	4.3	63.70	59.60
$Bi_{0.4}Sb_{1.6}Te_{3+x}$ x=0.24	90	30	30	1.0	61.53	61.37
			45	1.5	63.48	59.27
			60	2.0	63.34	60.34
			100	3.0	63.90	61.13
			135	4.0	62.68	59.48
			165	5.0	63.69	60.73

And the film growth process is highly repeatable, we have prepared several such high oriented samples. In the original manuscript, we have shown two samples of N-type Bi_2Te_{3+x}(x=0.17#1 and x=0.17#2) samples with almost same TE properties. We also check the repeatability for other different compositions including N-type and P-type samples as shown below. All the results showed very high consistent between different batch of both P-type and N-type samples.

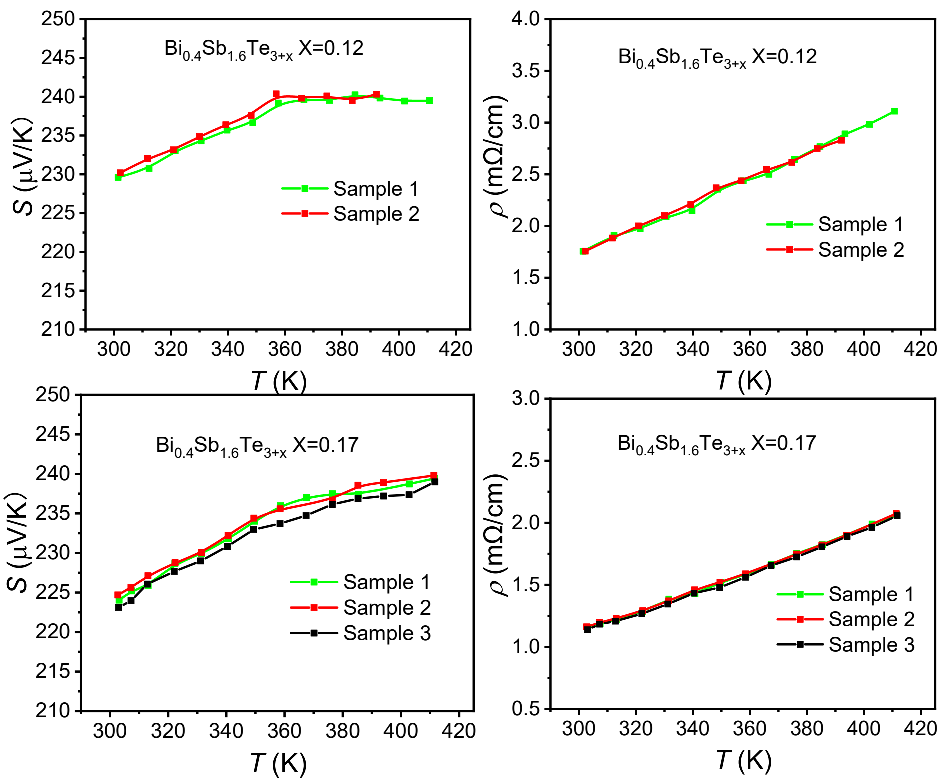


Fig. R3. Electrical properties repeatability for different p-type Bi_2Te_3-based films.

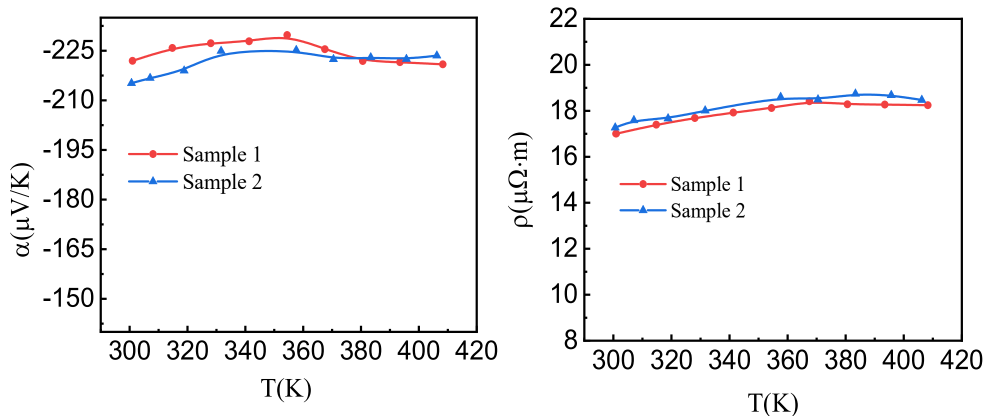


Fig. R4. Electrical properties repeatability for different n-type Bi_2Te_{3+x}(x=0.12) films.

(3) The microstructural analysis, while thorough, could be enhanced by a deeper exploration of how the observed microstructural features specifically influence phonon scattering and thermal conductivity. Correlating these structural properties with mechanical stability under operational stress conditions would also offer valuable insights.

Response: Thanks for the valuable suggestion.

As discussed in the manuscript, the Liquid-Te assistant growth process could induce tremendous of dislocation and stacking faults, which introduce severe stress into the lattice and scatter the phonon greatly. We carefully simulated the different phonon scattering effecton the lattice thermal conductivity in Debye model, including point defects, one dimensional defect dislocation and two dimensional defect stacking fault. As shown in Fig. 5 and Table S4-S5, the point defect scattering, and acoustic phonon scattering were close in $Bi_{0.4}Sb_{1.6}Te_{3+x}$(x=0 and 0.17) samples, the grain boundary scattering is weakened by the enlarged grains. Importantly, the dislocation and stacking faults scattering are greatly enhanced in x=0.17 samples. Therefore, the sample with high density of staggered layers exhibit very low thermal conductivity as shown in Fig. 5.

Different with previous study on single crystal nanosheet (Lu, et al, *Nature Nanotechnol.* 2023, 18**:**1281 – 128), the staggered layers is generated in a Te-rich annealing process. In principle, this microstructure is thermodynamic stable in current study. Of course, in the real Chip package structure, the TIM layers is under a operation stress condition, the stability under the real conditions including in situ thermal and stress conditions is important issue for future application.

Reviewer #2 (Remarks to the Author):

How to improve the texture degree of the (00*l*) plane is a very important issue for Bi_2Te_3-based thermoelectric films. This work intelligently proposed an extra-Te-assisted method to achieve high (00*l*) orientation of both p- and n-type Bi_2Te_3-based films and profoundly discovered the mechanics of grain growth with extra Te by the chemical bonding theory. This method produces the orientation of polycrystal film to be comparable with those of single sheets, which gets effect instantly in the improvement of the electrical properties. On the other hand, the extra Te in the film provided by this method induces rich microstructures, such as dislocations and stacking faults, which can strongly depress the thermal conductivity. Finally, exciting performances of both Bi_2Te_3-based films (zT=1.5) and the film device (ΔT=~8.2 K) have been achieved. Both the result and analysis are solid and organized well. Before the manuscript is accepted, there are still some questions that need to be solved.

(1) The high texture degree of the (00*l*) plane produces high carrier mobility by this extra-Te-assisted method, but this advantage has been not strongly illustrated for the lack of necessary comparison.

Response: Thanks. The comparison of carrier mobility is listed for samples prepared in this work and reported result, the result is added as Fig. 4 and Fig. S13 in the Supporting Information. The mobility for P-type $Bi_{0.4}Sb_{1.6}Te_3$ film were enhanced from 213 cm^2/V-s in x=0 sample to 300 cm^2/V-s in x=0.17 sample, and for N-type Bi_2Te_3, the mobility is enhanced from 93 cm^2/V-s to 120 cm^2/V-s, as shown in the following Fig. R4.We also modified the Fig.S14 to emphasize this enhancement.

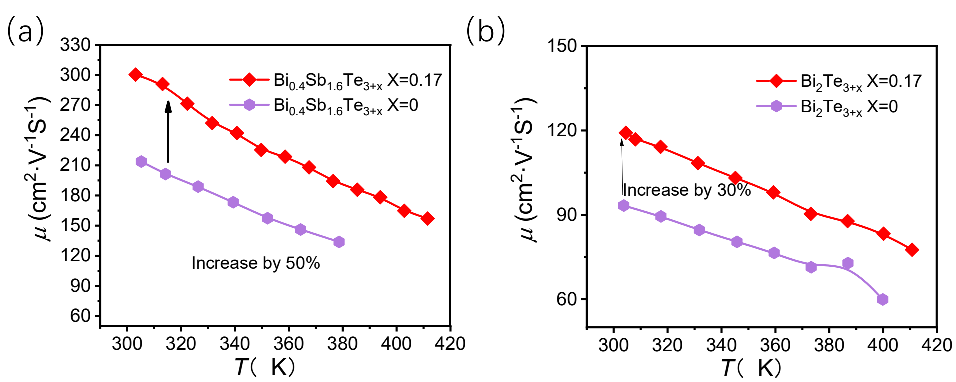


Fig. R5. The temperature dependent hall mobility for (a) P-type $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (b) and N-type Bi_2Te_{3+x} (x=0 and 0.17)

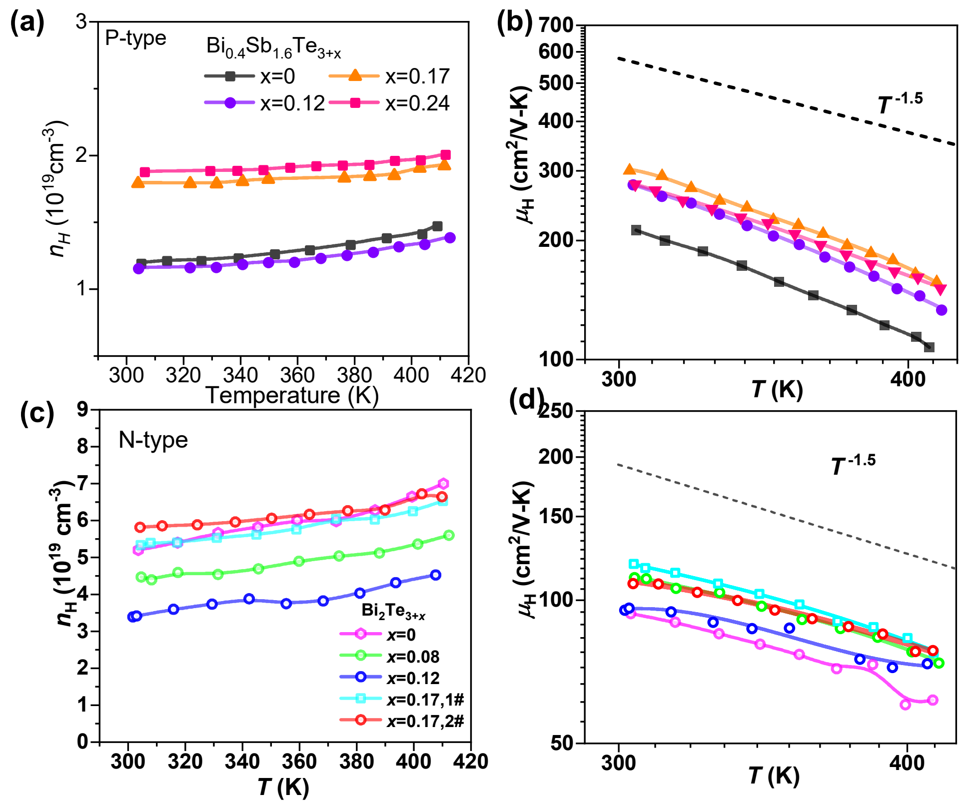


Fig. S14. **Electrical properties for p- and n-type Bi_2Te_3-based samples.** The temperature dependent hall carrier concentration and hall mobility for *p*-type $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0, 0.12, 0.17 and 0.24) (a~b) and *n*-type Bi_2Te_{3+x} (x=0, 0.08, 0.12 and 0.17) (c~d) films in this work. The $T^{-1.5}$ temperature dependence of hall mobility indicated the dominated acoustic phonon scattering in this work.

(2) The extra Te can induce the Te–(BiSb)–Te layers and Te–(BiSb)–Te–(BiSb)–Te–(BiSb)–Te seven-layers, but it little affects the carrier mobility, can the author provide a relevant explanation? Besides, is there evidence to verify the similar compositions of x=0.17 and x=0.24 after annealing?

Response: Thanks. As suggested by the reviewer, the Te –(BiSb)-Te layers and Te-(BiSb)-Te-(BiSb)-Te-(BiSb)-Te seven layer has suppressed the lattice thermal conductivity but the microstructure has little effect on the carrier mobility. According to Fig. 4d, the carrier mobility of the carriers is determined by the scattering mechanism and relaxation time of carriers. The $T^{-1.5}$ temperature dependence of carrier indicated the phonon to phonon scattering dominated, then the temperature dependent mean free length of the carriers can be obtained as below, which are much larger than layer space of Bi_2Te_3. Although extra Te induce high density of staggered layers, these layers still are in the same direction with the *ab* plane of unit cell, which is the predominant direction of thermoelectric performance. In this direction, the dimension of stacking faults would be not larger than several atomic layer spaces, which is much smaller than the mean free length of carriers. Therefore, the in-plane transport of carriers is weakly affected.

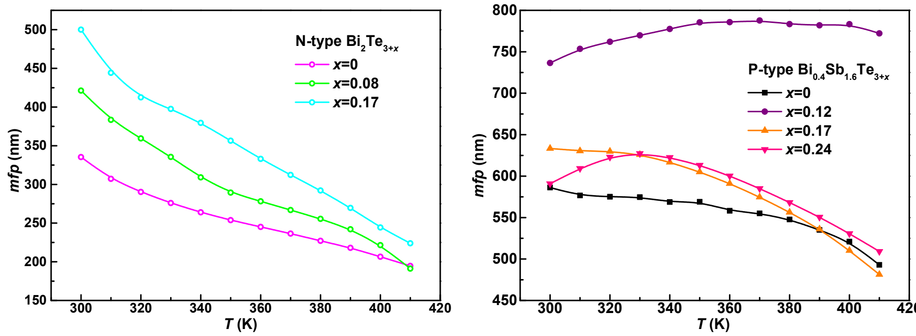
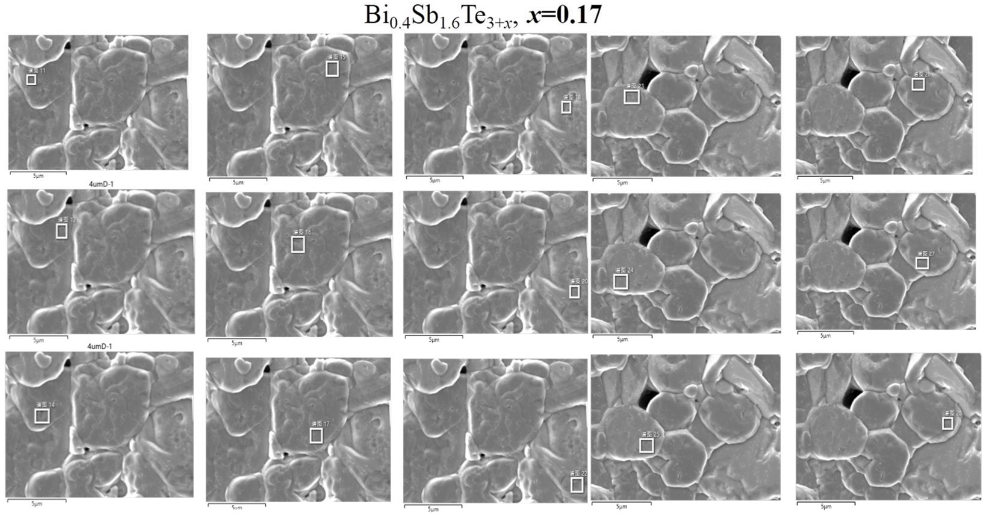


Fig. R6. The temperature dependent mean free length of the carriers for $Bi_{0.4}Sb_{1.6}Te_{3+x}$ and Bi_2Te_{3+x} sample.

The elements composition of $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.17 and x=0.24) samples were investigated by characterizing more than 15 points for each sample as shown as following. It can be seen that, the average composition of these two samples are quite close after annealing considering the testing accuracy.

$Bi_{0.4}Sb_{1.6}Te_{3+x}$, $\boldsymbol{x}$**=0.17**



$Bi_{0.4}Sb_{1.6}Te_{3+x}$, x=0.24

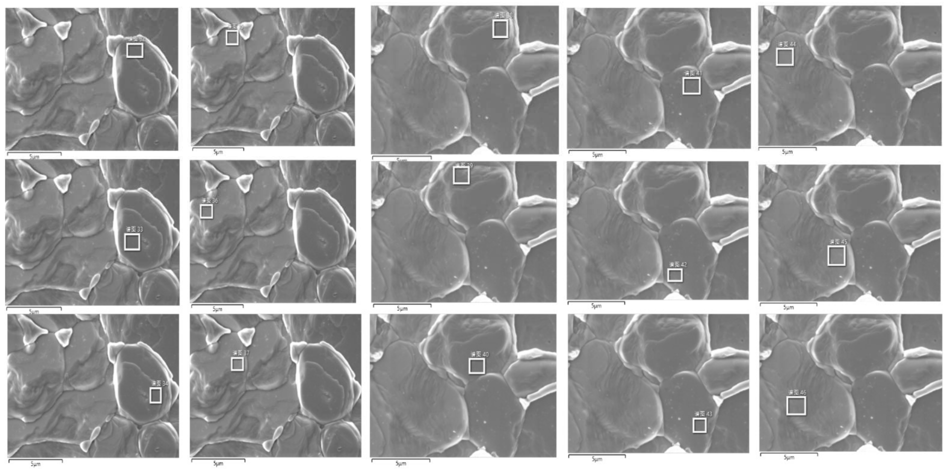


Fig. R7. The SEM image for the EDS characterization of $Bi_{0.4}Sb_{1.6}Te_3$ (x=0.17 and x=0.24)

Table R2. the EDS results of $Bi_{0.4}Sb_{1.6}Te_{3+x}$ (x=0.17 and x=0.24)

Samples	x=0.17			x=0.24		
Positions	Bi(at%)	Sb(at%)	Te(at%)	Bi(at%)	Sb(at%)	Te(at%)
1	8.37	31.82	59.81	8.17	32.53	59.31
2	7.83	32.1	60.07	8.18	32.37	59.45
3	7.82	31.97	60.22	8.06	32.27	59.67
4	8.4	32.29	59.3	8.73	31.44	59.83
5	8.45	32.3	59.25	8.77	32.46	58.77
6	8.42	32.61	58.97	8.32	32.3	59.38
7	7.6	31.73	60.67	8.18	32.43	59.39
8	7.71	32.28	60.02	8.31	31.93	59.75
9	7.5	32.27	60.23	8.03	32.56	59.41
10	8.69	32.12	59.19	8.41	32.02	59.57
11	8.97	32.06	58.97	8.21	31.9	59.88
12	8.57	32.3	59.12	8.16	31.82	60.02
13	7.97	32.37	59.66	8.56	32.14	59.3
14	8.48	32.45	59.07	8.44	32.62	58.94
15	7.75	32.32	59.93	8.36	32.1	59.5
Average	8.17	32.2	59.63	8.32	32.19	59.48

(3) The words in Fig.1a are in disorder, please rearrange the words properly.

Response: Thanks. Sorry for the format error in the PDF conversion. Fig.1a is replaced right now.

(4)"... that the growth rate along (00l) planes is almost ten times higher than that of vertical (00l) planes, and also about 3 times higher than that along (015) planes, as shown in Figure 2...” the expression of (00l) planes and (015) planes should be corrected.

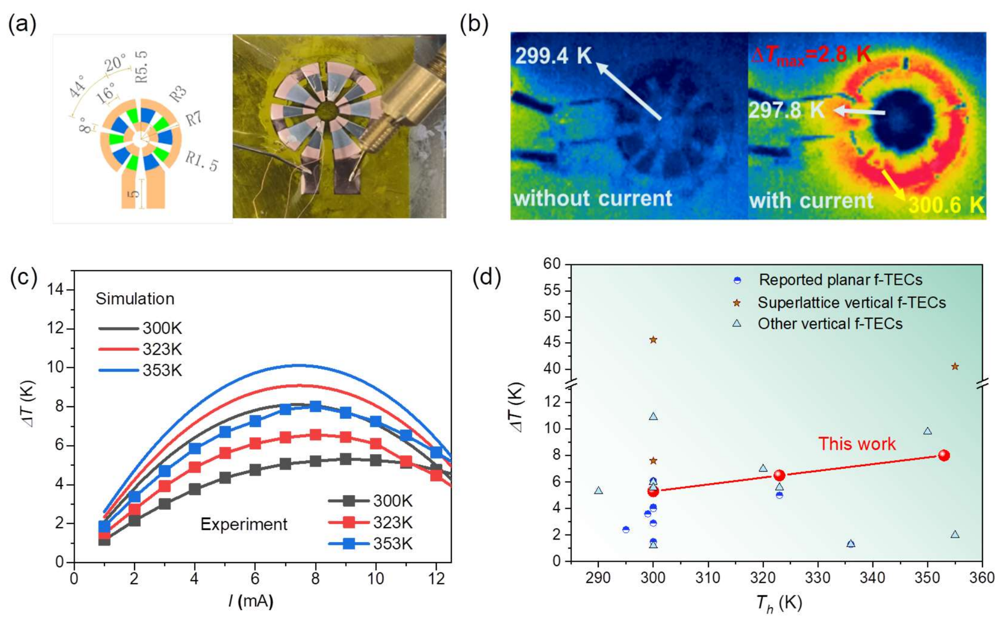
Response: Thanks. Sorry for the mistake, the expressions have been revised.

(5) The EDS in Fig. S7 shows Te second phase between grain boundaries, however, the authors claimed extra Te has evaporated after annealing at the last of page 4, please explain the inconsistence.

Response: Thanks. Sorry for the inaccurate expression. Actually, although final annealing temperature is higher than the Te melting point, there were trace of Te residual on the grain boundary. Nevertheless, the Te content inside the grains are quite close between different samples, since the Te in the lattice were saturated as shown in Fig. R6. The sentences at the last of Page 4 have been revised as “...but the component inside of $Bi_{0.4}Sb_{1.6}Te_3$ and Bi_2Te_3 grains are close to the stoichiometric ratio. ”

(6) The format of the figure 6 should be consistent with other figures.

Response: Thanks. Sorry for the inconsistence, Fig.6 has been replaced as follows:



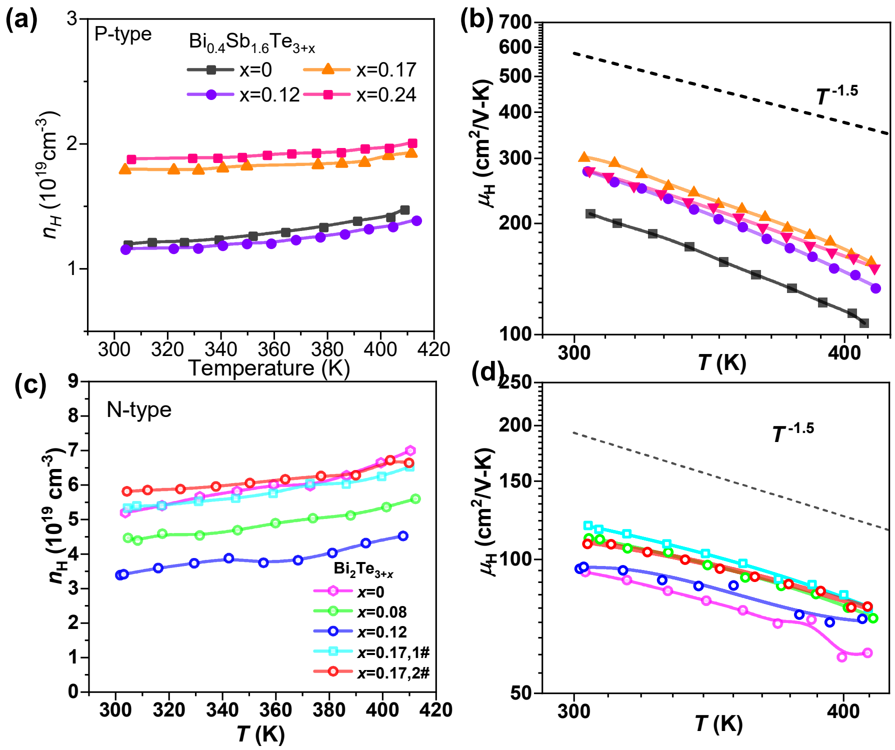
Reviewer #3 (Remarks to the Author):

In this work, highly (00l)-oriented Bi_2Te_3 films, comparable to single crystals, were obtained using a liquid Te-assisted growth method. The high mobility resulting from the orientation, combined with low lattice thermal conductivity due to Te-induced staggered stacking faults, led to in-plane zT values of ~1.53 for P-type $Bi_{0.4}Sb_{1.6}Te_3$ and ~1.10 for N-type Bi_2Te_3 films. Planar f-TEC devices based on these films achieved a temperature reduction of ~8.2 K in hot spot elimination experiments, demonstrating the potential of this method for producing oriented planar Bi_2Te_3 films and for f-TEC device design, with important implications for next-generation thermal management in integrated circuits. Therefore, I recommend its publication in Nature Communication after addressing the following:

Response: Thanks for the recommendation.

1. Please carefully check the accuracy of all images and their annotations throughout the text. For example, the annotation for S12 in Figure 3 does not match the image; the meanings of x=0.17 1# and x=0.17 2# in Figure S13 are not explained in the text.

Response: Sorry for the mistake. The data have already been double checked and revised. The annotations x=0.17 #1 and x=0.17 #2 is represent two parallel samples, and the data of two samples show good consistency in electrical properties in the Fig.S13 and Fig. S14. And the carrier concentration and mobility data of sample Bi_2Te_{3+x} (x=0.08) is also added in Fig. S14.



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2. Some typos: line 51, 2, 5; line 66 powder factor; line 94, comparasion, leaser, etc

Response: Thanks. Sorry for the typos, and they are already been revised.

3. Please carefully check if the data mentioned in the text matches the figures. For example, verify whether the value of 0.08 in "F jumps from 0.08 for $x = 0$ to 0.84 for $x = 0.24$" on lines 127-128 corresponds correctly with the data shown in the figures.

Response: Sorry for the mistakes. The XRD and orientation data of P-type x=0.24 sample were depicted in Fig.S6. We have modified the text in the manuscript into "...from 0.08 for x = 0 to 0.68 fo x = 0.17, and then to 0.84 for x = 0.24, which is comparable to the zone-melting crystals (Fig. 2d and Fig. S6)."

4. Please provide the manufacture information of Magnetron sputtering device. And how long it takes to deposit the BT films?

Response: Thanks. The manufacture process of BT films is added in the Method section.

5. What is the reason for the gap between the experiment and the simulated values in Figure 6? How long could the high-record delta T keep when the laser and TEC is on together?

Response: Thanks.
The gap maybe come from two reasons. The first reason is the uncertainty of thermal conductivity of PI substrate, and the bonding state between films and substrate, the other reason is the testing condition, since the simulation is carried out in an ideal insulating environment, which is hardly achieved in the real testing experiment due to the heat loss from residual air, thermal couple wires, current wire, and so on, as shown in the Figure 6.
As shown in Fig.1 and Fig. S16, the maximum delta T between the final temperature and the highest temperature only laser on was acquisited after the whole system entering the stable state, so if the power of laser and TEC is not changed, the delta T could keep up for ever.

Dear Editors and Reviewers:

Thank you for your letter and for the reviewers' comments concerning our manuscript entitled "*Oriented Bi2Te3-based films enabled high performance planar thermoelectric cooling device for hot spot elimination*" (ID: NCOMMS-24-53304A). Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction which we hope meet with approval. Please find below our detailed responses as well as our revisions, where the modified parts are highlighted in the manuscript (marked in blue font).

Reviewer #1 (Remarks to the Author):

The authors have satisfactorily addressed the issues and concerns I raised in my initial review. I think the paper can be published now

Response: Thank you for your recognition of this work. Thank you again for your important suggestions to improve this work.

Reviewer #2 (Remarks to the Author):

I believe the author has addressed my concerns, and the article is now logically sound and acceptable.

Response: Thanks for the concerns which you listed to make this work more prefect, and we are pleasant to work it out for addressing your concerns.

Reviewer #3 (Remarks to the Author):

Publish as is

Response: Thank you very much for your key questions that make this work more reasonable and logical. Thank you for your recognition.

Thank the editor and reviewers again for their time and thoughtful comments. We hope that the revised manuscript would be more suitable for the publication in "***Nature Communication***"

Sincerely,

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