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The pressure-induced structural transitions, alloying and superconductivity in topological insulators Bi_2Te_2Se and Bi_2Se_2Te

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Fig. S1 Comparison of experimental and simulated XRD patterns for Bi_2Te_2Se at 47.3 GPa.

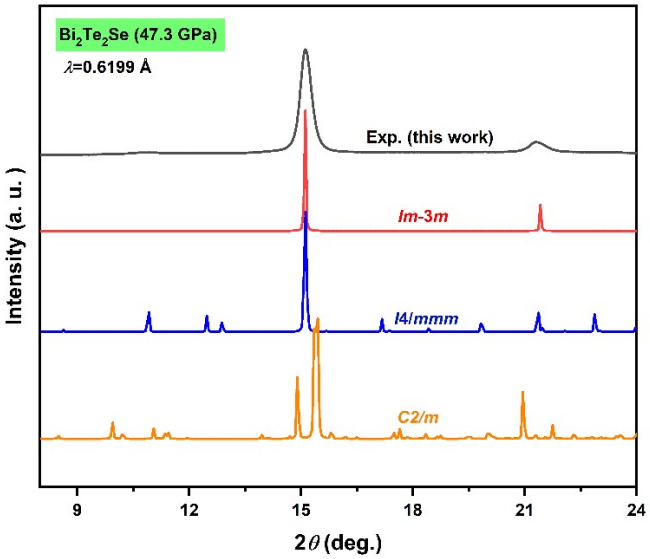

Fig. S2 Experimental and fitted (red line) XRD patterns for phase IV of Bi_2Se_2Te at 36.3 GPa, which are simulated by using the **(a)** *C*2/*m* (LeBail fitting), **(b)** BCT (Rietveld refinement) and **(c)** disordered BCC structures (Rietveld refinement), respectively.

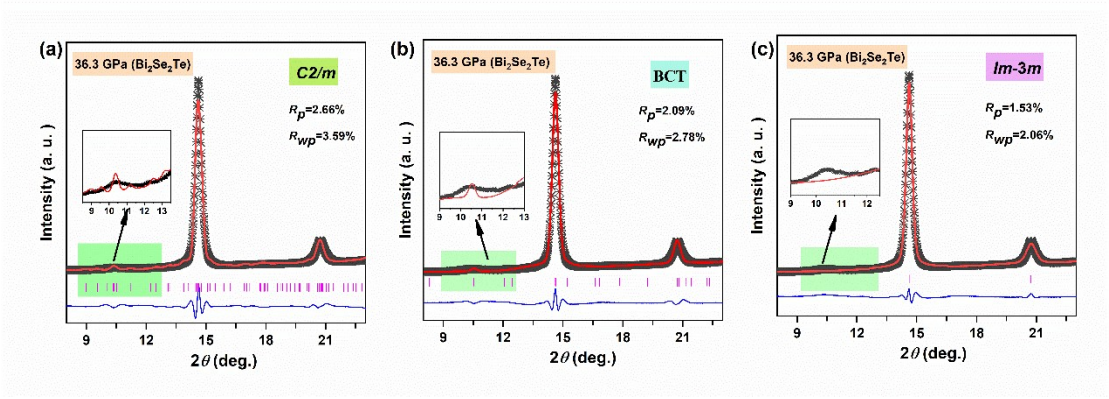

Table S1. Lattice parameters and atomic coordinates of phase I and II of Bi_2Se_2Te and Bi_2Te_2Se, respectively.

Lattice params	atoms	x	y	z
Phase I (*R*-3*m*) 0.3 GPa for Bi_2Te_2Se				
a=4.2315(5) Å	Bi1 (6c)	0	0	0.3988(6)
c=29.4592(1) Å	Te1 (6c)	0	0	0.1972(1)
	Se3 (3a)	0	0	0
Phase II (*C*2/*m*) 12.0 GPa for Bi_2Te_2Se				
	Bi1 (4i)	0.3149(19)	0	0.3351(19)
a=14.1857(14) Å	Bi2 (4i)	0.4957(20)	0	0.2706(18)
b=3.8780(11)	Te1 (4i)	0.0610(32)	0	0.9207(27)
c=16.8840(22) Å	Te2 (4i)	0.8601(27)	0	0.5128(23)
β=147.504(2) °	Se1 (4i)	0.6590(48)	0	0.0689(31)
Phase I (*R*-3*m*) 0.4 GPa for Bi_2Se_2Te				
a=4.2026(2) Å	Bi1 (6c)	0	0	0.3973(4)
c=29.1356(3) Å	Se1 (6c)	0	0	0.2101(3)
	Te3 (3a)	0	0	0
Phase II (*C*2/*m*) 16.4 GPa for Bi_2Se_2Te				
	Bi1 (4i)	0.3365(12)	0	0.3047(18)
a=14.1787(42) Å	Bi2 (4i)	0.5063(15)	0	0.2655(13)
b=3.8180(37)	Se1 (4i)	0.1124(19)	0	0.8276(22)
c=16.5224(22) Å	Se2 (4i)	0.8531(23)	0	0.4560(34)
β=147.936(6) °	Te1 (4i)	0.7209(21)	0	0.0881(18)

Table S2. The unit cell results versus pressure of phase I, II and III for Bi_2Te_2Se *via* Le Bail fitting method.

Phase I (*R*-3*m*)			
***P* (GPa)**	***a*/Å**	***c*/Å**	***V*/ Å^3**
0.3	4.2315(5)	29.4592(1)	456.991(3)
1.4	4.2070(1)	29.1635(2)	447.008(4)
2.0	4.1935(2)	28.9970(5)	441.608(2)
2.6	4.1815(5)	28.8601(4)	437.010(8)
4.2	4.1532(6)	28.6029(1)	427.275(7)
5.6	4.1262(3)	28.4408(1)	419.361(4)
6.7	4.1106(2)	28.3669(4)	415.101(5)
7.6	4.0962(2)	28.2805(2)	410.958(6)
9.0	4.0755(1)	28.1766(3)	405.304(2)
10.2	4.0555(6)	28.0905(2)	400.125(9)

Phase II (*C*/2*m*)					
***P* (GPa)**	***a*/Å**	***b*/Å**	***c*/Å**	***β*/°**	***V*/ Å^3**
10.2	14.3526(12)	3.8881(15)	17.0710(16)	147.656(1)	509.661(1)
12.0	14.1857(14)	3.8780(11)	16.8840(22)	147.504(2)	499.003(6)
13.2	14.1407(12)	3.8736(13)	16.7543(12)	147.368(1)	494.879(6)
15.3	14.0752(16)	3.8681(12)	16.6034(33)	147.320(2)	488.101(7)
17.2	14.0176(11)	3.8632(20)	16.4528(23)	147.286(1)	481.522(2)
19.5	13.9856(21)	3.8577(24)	16.2586(27)	147.231(2)	474.781(3)
Phase III (*C*/2*c*)					
19.5	9.5361(11)	6.4152(35)	9.8215(35)	130.450(1)	457.223(8)
24.8	9.3861(23)	6.3150(21)	9.7253(24)	130.200(1)	440.290(6)
28.4	9.3113(21)	6.2658(13)	9.6885(37)	130.066(7)	432.589(3)
32.1	9.2456(18)	6.2237(15)	9.6420(28)	130.035(8)	424.801(1)
36.7	9.1782(17)	6.1811(26)	9.5934(11)	130.015(2)	416.827(2)
38.9	9.1535(12)	6.1636(27)	9.5756(27)	129.995(5)	413.886(7)

Table S3. The unit cell results versus pressure of phase I and II for Bi_2Se_2Te *via* Le Bail fitting method.

Phase I (*R*-3*m*)			
***P* (GPa)**	**a/Å**	**c/Å**	**V/ Å^3**
0.4	4.2026(2)	29.1356(3)	445.647(6)
1.8	4.1779(1)	28.7677(2)	434.862(7)
3.4	4.1343(2)	28.3079(7)	419.027(8)
5.5	4.0833(3)	27.9014(5)	402.883(7)
6.8	4.0585(1)	27.7486(3)	395.825(5)
7.9	4.0361(2)	27.6595(6)	389.210(2)
9.1	4.0142(5)	27.5484(8)	384.437(7)
10.7	3.9983(9)	27.4208(3)	379.631(6)

Phase II (*C*/2*m*)					
***P* (GPa)**	***a*/Å**	***b*/Å**	***c*/Å**	***β*/°**	***V*/ Å^3**
10.7	14.2253(52)	3.91652(14)	16.7852(17)	148.103(3)	494.136(8)
13.2	14.1988(21)	3.8622(27)	16.6192(43)	148.005(2)	482.885(4)
16.4	14.1787(42)	3.8180(37)	16.5224(22)	147.936(6)	474.822(5)
18.9	14.1681(23)	3.7721(18)	16.4178(31)	147.909(8)	466.147(8)
23.4	14.1493(28)	3.7045(25)	16.2734(22)	147.872(1)	453.505(7)
26.8	14.1339(31)	3.6535(43)	16.1748(20)	147.745(5)	445.757(5)

Table S4. Lattice parameters and atomic coordinates of phase IV of Bi_2Se_2Te at 36.3 GPa, which are simulated by using the body-centered tetragonal (BCT), 9/10-fold monoclinic, and disordered body-centered cubic (*Im*-3*m*) structures

Lattice params	atoms	x	y	z
Phase IV (*C*2/*m*) 36.3 GPa				
	Bi1 (4i)	0.3995	0	0.6081
a=14.7858 Å	Bi2 (4i)	0.1961	0	0.8060
b=4.9031 Å	Se1 (4i)	0.3998	0	0.1028
c=5.9465 Å	Se2 (2a)	0	0	0
β=104.809°	Se3 (2c)	0	0	0.5
	Te (4i)	0.7978	0	0.6940
Phase IV (BCT) 36.3 GPa				
	Bi (4e)	0	0	0.4105(3)
a=3.4518(5) Å	Se (4e)	0	0	0.2039(2)
c=17.1806(8) Å	Te (2a)	0	0	0
Phase IV (*Im*-3*m*) 36.3 GPa				
a=3.4298(1) Å	Bi/Te/Se (2a)	0	0	0

Table S5. The unit cell results versus pressure of phase IV for Bi_2Te_2Se and Bi_2Se_2Te obtained from Rietveld refinements/ Le Bail fitting using different space groups.

Phase IV (*Im*-3*m*) of Bi_2Te_2Se (Rietveld method)			
***P* (GPa)**	***a*/Å**	***V*/ Å^3**	***Rp*/*Rwp* (%)**
24.8	3.4541(2)	41.212(3)	3.12/4.55
28.4	3.4420(1)	40.779(4)	2.79/3.86
32.1	3.4266(3)	40.236(8)	2.62/5.25
36.7	3.4075(2)	39.565(5)	2.52/3.60
38.9	3.3971(8)	39.205(4)	1.59/2.64
42.7	3.3802(7)	38.621(6)	1.43/2.32
45.3	3.3705(3)	38.290(1)	1.65/2.58
47.3	3.3601(6)	37.935(1)	1.76/2.38
50.7	3.3442(7)	37.400(2)	1.25/3.05
55.1	3.3208(5)	36.621(3)	1.03/1.93
60.5	3.2982(1)	35.879(1)	0.94/1.46
Phase IV (*Im*-3*m*) of Bi_2Se_2Te (Rietveld method)			
26.8	3.4664(2)	41.653(3)	2.57/3.95
30.6	3.4488(2)	41.021(7)	2.49/3.73
32.5	3.4398(3)	40.702(4)	2.32/3.41
36.3	3.4298(9)	40.347(1)	1.53/2.06
37.8	3.4259(2)	40.209(2)	2.53/3.72
41.3	3.4147(1)	39.816(5)	2.15/3.26
46.8	3.3998(6)	39.300(6)	1.74/2.36
49.3	3.3902(5)	38.966(5)	2.07/3.08
52	3.3794(4)	38.593(3)	2.07/3.44
55.9	3.3663(2)	38.145(2)	1.85/2.06
61.5	3.3446(7)	37.416(1)	0.91/1.50
66.3	3.3317(5)	36.984(1)	1.40/1.96

Phase IV (BCT) of Bi_2Te_2Se (Rietveld method)				
***P* (GPa)**	***a*/Å**	***c*/Å**	***V*/ Å^3**	***Rp*/*Rwp*(%)**
24.8	3.5295(1)	17.5683(5)	218.855(8)	3.68/4.55
28.4	3.4987(2)	17.4054(1)	213.058(5)	3.51/4.25
32.1	3.4737(2)	17.2929(7)	208.666(9)	3.33/4.08
36.7	3.4429(6)	17.1711(2)	203.539(8)	3.05/4.13
38.9	3.4133(1)	17.0442(5)	198.575(2)	2.78/3.77
42.7	3.3862(8)	16.9230(2)	194.045(6)	2.55/3.63
45.3	3.3757(2)	16.8155(8)	191.618(4)	2.15/3.42
47.3	3.3650(6)	16.7283(8)	189.418(4)	2.01/3.10
50.7	3.3440(3)	16.4838(1)	184.327(3)	2.08/3.45
55.1	3.3226(4)	16.3796(5)	180.834(7)	2.05/3.23
60.5	3.2880(8)	16.3068(4)	176.298(1)	2.11/3.30
Phase IV (BCT) of Bi_2Se_2Te (Rietveld method)				
26.8	3.4928(1)	17.2954(2)	210.997(2)	4.13/5.67
30.6	3.4769(7)	17.2511(2)	208.545(7)	3.88/4.35
32.5	3.4642(3)	17.2205(3)	206.657(5)	3.52/4.78
36.3	3.4541(8)	17.1907(5)	205.098(7)	2.09/2.78
37.8	3.4402(9)	17.1646(8)	203.142(8)	3.05/4.18
41.3	3.4187(5)	17.1297(4)	200.203(3)	2.55/3.67
46.8	3.3990(6)	17.0659(6)	197.165(2)	2.16/3.08
49.3	3.3901(2)	17.0131(3)	195.527(1)	2.55/3.56
52	3.3807(4)	16.9511(2)	193.736(2)	2.18/3.27
55.9	3.3601(5)	16.8891(1)	190.682(5)	2.05/3.13
61.5	3.3398(6)	16.8070(5)	187.469(4)	2.54/3.81
66.3	3.3119(7)	16.7158(5)	183.350(7)	2.17/3.42

Phase IV (*C*/2*m*) of Bi_2Te_2Se (Le Bail fitting)						
***P* (GPa)**	***a*/Å**	***b*/Å**	***c*/Å**	***β*/°**	***V*/ Å^3**	***Rp*/*Rwp*(%)**
24.8	15.5978(15)	5.2583(21)	6.3211(35)	106.124(8)	498.050(2)	5.68/7.55
28.4	15.4236(22)	5.2431(53)	6.2856(26)	105.985(5)	488.649(2)	6.52/7.88
32.1	15.3629(24)	5.2314(27)	6.2398(35)	105.841(3)	482.451(7)	4.88/6.12
36.7	15.2915(25)	5.2222(16)	6.2058(23)	105.691(5)	477.108(8)	5.13/6.33
38.9	15.1256(27)	5.2093(22)	6.1877(45)	105.551(2)	469.709(5)	5.44/7.04
42.7	14.9942(25)	5.2025(17)	6.1742(15)	105.328(1)	464.506(4)	4.32/5.16
45.3	14.8653(27)	5.1967(22)	6.1549(16)	105.162(2)	458.923(2)	3.55/4.67
47.3	14.7534(32)	5.1923(41)	6.1327(18)	104.986(8)	453.821(2)	2.14/3.23
50.7	14.5781(12)	5.1797(16)	6.1227(31)	104.694(9)	447.207(3)	2.33/3.68
55.1	14.3321(22)	5.1454(34)	6.1044(54)	104.254(7)	436.315(1)	2.55/3.59
60.5	13.8806(13)	5.0771(25)	6.0786(18)	103.633(3)	416.318(7)	2.22/3.49
Phase IV (*C*/2*m*) of Bi_2Se_2Te (Le Bail fitting)						
26.8	15.7369(21)	5.2770(3)	6.3990(34)	105.356(5)	512.425(3)	4.58/5.63
30.6	15.4421(24)	5.1861(31)	6.2915(25)	105.241(2)	486.163(2)	4.12/5.27
32.5	15.1915(34)	5.0359(11)	6.0942(34)	105.115(3)	450.093(8)	3.55/4.21
36.3	14.7858(13)	4.9031(21)	5.9465(22)	104.809(8)	416.779(9)	2.66/3.59
37.8	14.6585(22)	4.8014(41)	5.8500(32)	104.685(8)	398.280(5)	2.45/3.54
41.3	14.4569(37)	4.7261(12)	5.7838(18)	104.482(1)	382.625(6)	2.59/3.25
46.8	14.2985(34)	4.6126(22)	5.7193(19)	104.256(3)	365.593(2)	2.42/3.27
49.3	14.1394(28)	4.5074(15)	5.6626(9)	104.210(6)	349.851(5)	3.11/4.33
52	14.0234(16)	4.4278(34)	5.6079(12)	104.185(9)	337.600(7)	3.05/4.18
55.9	13.8842(13)	4.3410(18)	5.5516(32)	104.056(5)	324.590(3)	2.58/3.31
61.5	13.7192(17)	4.2456(13)	5.5027(25)	103.987(2)	311.009(1)	2.66/3.78
66.3	13.5562(12)	4.1258(12)	5.4568(47)	103.882(5)	296.285(4)	2.71/3.45