

Origin of the concentration-dependent effects of N on the stability and electrical resistivity in polycrystalline $\text{Ge}_1\text{Sb}_2\text{Te}_4$

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1. Statistics for bulk $\text{Ge}_1\text{Sb}_2\text{Te}_4$

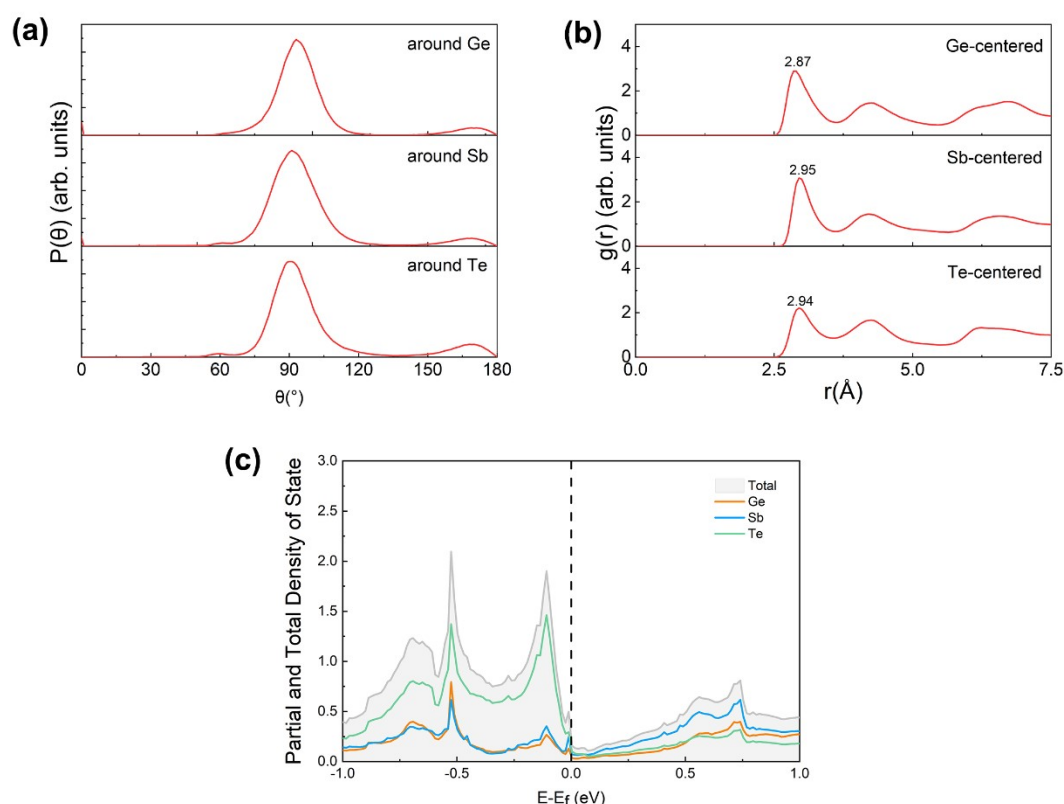


Fig. S1. Statistics for bulk $\text{Ge}_1\text{Sb}_2\text{Te}_4$ cell: (a) partial pair correlation functions (b) bond-angle distribution (c) partial and total density of states

2. N doped $\Sigma 3$ (111) $\text{Ge}_1\text{Sb}_2\text{Te}_4$ polycrystalline structures

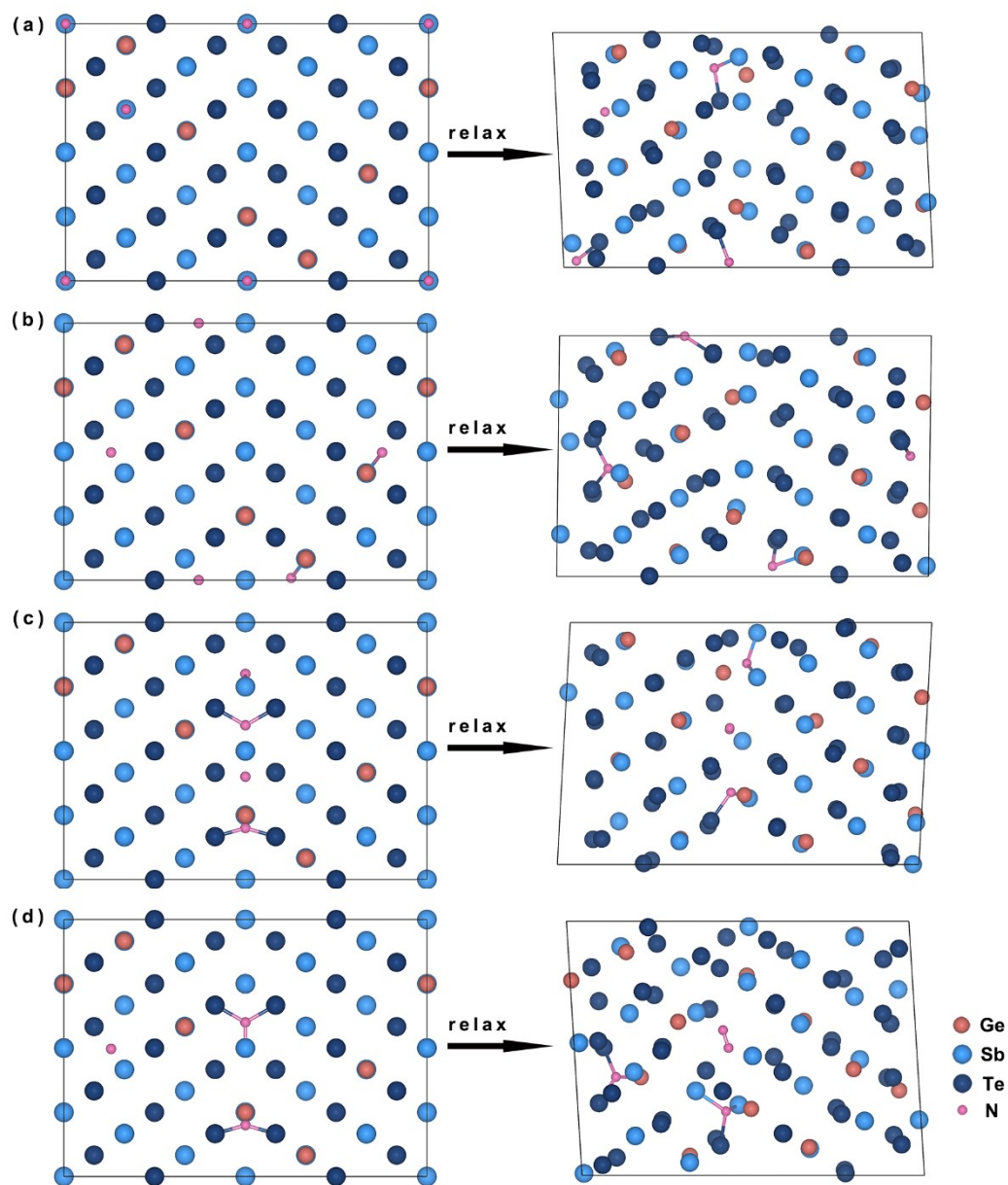


Fig. S2. The initial and relaxed $\Sigma 3(111)$ grain boundary structures for 4.55 at. % nitrogen doped GST124 cells: (a) vacancy doping (4N-v4-4a) (b) interstitial doping (4N-i4-4a) (c) boundary doping(4N-b4-2a1m) (d) 4N-b3i1-2a1m

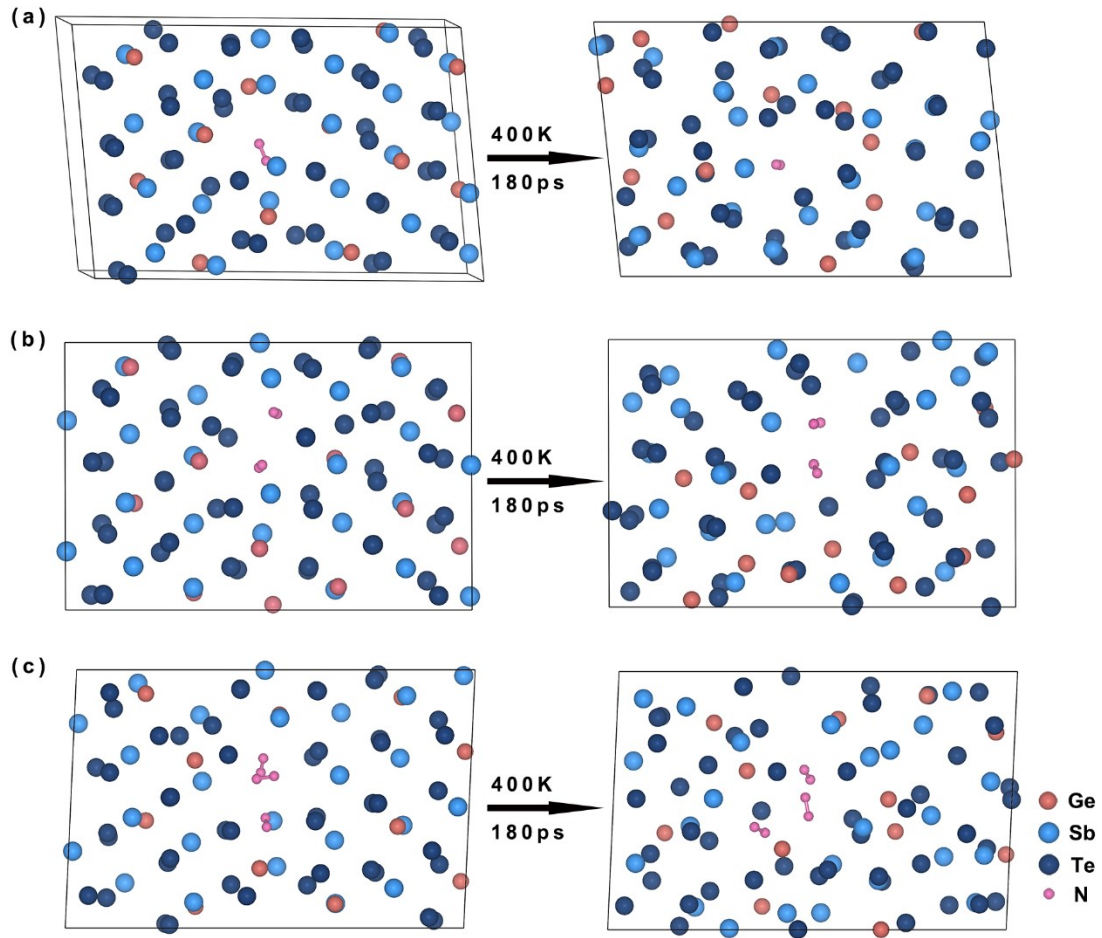


Fig. S3. GST124 $\Sigma 3(111)$ grain boundary structures with nitrogen molecule solely doped before and after 400K annealing: (a) 2N-b2-1m (b) 4N-b4-2m (c) 6N-b6-3m. The distance of the N_2 molecules are 3.13 Å in 4N-b4-2m and 3.15 Å/3.24 Å/3.68 Å for in 6N-b4-3m.

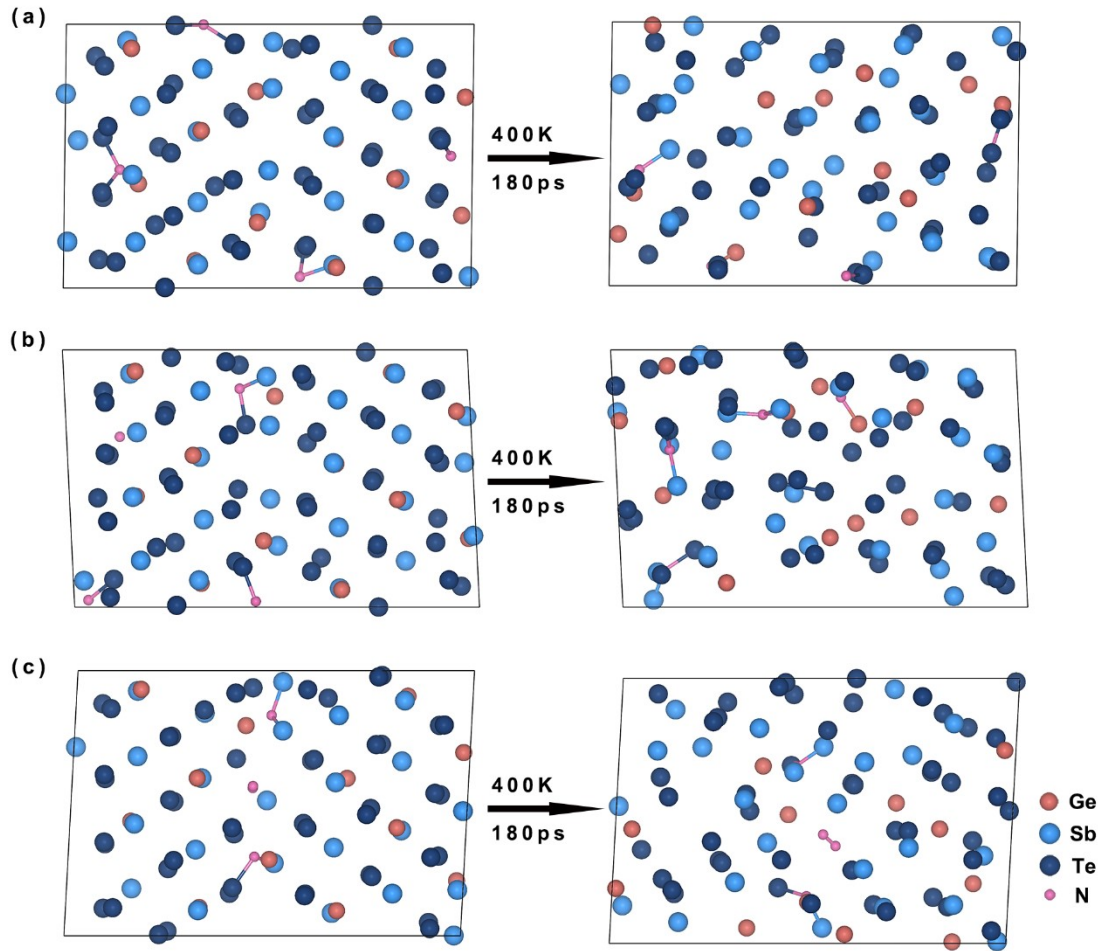


Fig. S4. GST124 $\Sigma 3(111)$ grain boundary structures with nitrogen molecule and nitride complex doped before and after 400K annealing: (a) 4N-i4-4a (b) 4N-v4-4a (c) 4N-b4-2a1m, nitride complex will stay stable after annealing instead of forming nitrogen molecule.

3. POSCAR files of grain boundary models:

$\Sigma 3(111)$ grain boundary:

GST124-s3

1.0

14.696938	0.0000000000	0.0000000000
0.0000000000	8.485281	0.0000000000
0.0000000000	0.0000000000	20.784609

Sb	Ge	Te
24	12	48

Direct

0.416667	0.750000	0.166667
0.083333	0.750000	0.333333
0.916667	0.250000	0.166667
0.583333	0.250000	0.333333
0.250000	0.250000	0.500000
0.750000	0.750000	0.500000
0.166667	0.000000	0.166667
0.666667	0.500000	0.166667
0.333333	0.500000	0.333333
0.000000	0.500000	0.500000
0.833333	0.000000	0.333333
0.500000	0.000000	0.500000
0.333333	0.500000	0.666667
0.166667	0.000000	0.833333
0.000000	0.500000	0.000000
0.833333	0.000000	0.666667
0.666667	0.500000	0.833333
0.500000	0.000000	0.000000
0.083333	0.250000	0.666667
0.583333	0.750000	0.666667
0.416667	0.250000	0.833333
0.250000	0.750000	0.000000
0.916667	0.750000	0.833333
0.750000	0.250000	0.000000
0.083333	0.750000	0.666667
0.583333	0.250000	0.666667
0.416667	0.750000	0.833333
0.250000	0.250000	0.000000
0.916667	0.250000	0.833333
0.750000	0.750000	0.000000
0.416667	0.250000	0.166667
0.083333	0.250000	0.333333
0.916667	0.750000	0.166667
0.583333	0.750000	0.333333
0.250000	0.750000	0.500000
0.750000	0.250000	0.500000
0.333333	0.000000	0.083333
0.000000	0.000000	0.250000
0.833333	0.500000	0.083333
0.500000	0.500000	0.250000
0.166667	0.500000	0.416667
0.666667	0.000000	0.416667
0.083333	0.750000	0.083333
0.583333	0.250000	0.083333

0.250000	0.250000	0.250000
0.750000	0.750000	0.250000
0.416667	0.750000	0.416667
0.916667	0.250000	0.416667
0.083333	0.250000	0.083333
0.583333	0.750000	0.083333
0.250000	0.750000	0.250000
0.750000	0.250000	0.250000
0.416667	0.250000	0.416667
0.916667	0.750000	0.416667
0.333333	0.500000	0.083333
0.000000	0.500000	0.250000
0.833333	0.000000	0.083333
0.500000	0.000000	0.250000
0.166667	0.000000	0.416667
0.666667	0.500000	0.416667
0.166667	0.500000	0.583333
0.000000	0.000000	0.750000
0.666667	0.000000	0.583333
0.500000	0.500000	0.750000
0.333333	0.000000	0.916667
0.833333	0.500000	0.916667
0.166667	0.000000	0.583333
0.000000	0.500000	0.750000
0.666667	0.500000	0.583333
0.500000	0.000000	0.750000
0.333333	0.500000	0.916667
0.833333	0.000000	0.916667
0.416667	0.750000	0.583333
0.250000	0.250000	0.750000
0.083333	0.750000	0.916667
0.916667	0.250000	0.583333
0.750000	0.750000	0.750000
0.583333	0.250000	0.916667
0.416667	0.250000	0.583333
0.250000	0.750000	0.750000
0.083333	0.250000	0.916667
0.916667	0.750000	0.583333
0.750000	0.250000	0.750000
0.583333	0.750000	0.916667

$\Sigma 5(100)$ grain boundary:

GST124-s5

1.0

5.9923501015	0.0000000000	0.0000000000
0.0000000000	13.3992996216	0.0000000000
0.0000000000	0.0000000000	26.7985992432

Sb	Ge	Te
20	10	40

Direct

0.000000000	0.099999998	0.150000011
-0.000000000	0.700000014	0.049999999
0.000000000	0.500000000	0.250000000
0.000000000	0.300000021	0.449999979
0.000000000	0.899999957	0.350000007
0.500000000	0.199999996	0.049999999
0.500000000	0.000000000	0.250000000
0.500000000	0.600000043	0.150000011
0.500000000	0.399999993	0.350000007
0.500000000	0.799999986	0.449999979
-0.000000000	0.300000021	0.550000021
-0.000000000	0.899999957	0.649999957
0.000000000	0.500000000	0.750000000
-0.000000000	0.099999998	0.850000043
-0.000000000	0.700000014	0.950000014
0.500000000	0.099999998	0.600000043
0.500000000	0.700000014	0.700000014
0.500000000	0.300000021	0.799999986
0.500000000	0.899999957	0.899999957
0.500000000	0.500000000	0.000000000
0.500000000	0.300000021	0.199999996
0.500000000	0.099999998	0.399999993
0.500000000	0.899999957	0.099999998
0.500000000	0.700000014	0.300000021
0.500000000	0.500000000	0.500000000
0.500000000	0.799999986	0.550000021
0.500000000	0.399999993	0.649999957
0.500000000	0.000000000	0.750000000
0.500000000	0.600000043	0.850000043
0.500000000	0.199999996	0.950000014
0.500000000	0.099999998	0.150000011
0.500000000	0.700000014	0.049999999
0.500000000	0.500000000	0.250000000
0.500000000	0.300000021	0.449999979
0.500000000	0.899999957	0.350000007
0.500000000	0.399999993	0.099999998
0.500000000	0.199999996	0.300000021

0.500000000	0.000000000	0.500000000
0.500000000	0.799999986	0.199999996
0.500000000	0.600000043	0.399999993
-0.000000000	0.199999996	0.049999999
0.000000000	0.000000000	0.250000000
-0.000000000	0.600000043	0.150000011
0.000000000	0.399999993	0.350000007
0.000000000	0.799999986	0.449999979
-0.000000000	0.300000021	0.199999996
0.000000000	0.099999998	0.399999993
0.000000000	0.899999957	0.099999998
-0.000000000	0.700000014	0.300000021
0.000000000	0.500000000	0.500000000
0.500000000	0.300000021	0.550000021
0.500000000	0.899999957	0.649999957
0.500000000	0.500000000	0.750000000
0.500000000	0.099999998	0.850000043
0.500000000	0.700000014	0.950000014
0.500000000	0.600000043	0.600000043
0.500000000	0.199999996	0.700000014
0.500000000	0.799999986	0.799999986
0.500000000	0.399999993	0.899999957
0.500000000	0.000000000	0.000000000
-0.000000000	0.099999998	0.600000043
-0.000000000	0.700000014	0.700000014
-0.000000000	0.300000021	0.799999986
0.000000000	0.899999957	0.899999957
0.000000000	0.500000000	0.000000000
-0.000000000	0.799999986	0.550000021
0.000000000	0.399999993	0.649999957
0.000000000	0.000000000	0.750000000
0.000000000	0.600000043	0.850000043
-0.000000000	0.199999996	0.950000014