Electronic Supplementary Information

Revealing the origin of dislocations in $Pb_{1-x}Sb_{2x/3}Se$ ($0<x\leq0.07$)

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Formation energy calculations

In order to understand the stability of the nanostructures in the matrix, we need to calculate its formation energy (E_{Nano}^{form}) using the following formula,

$$E_{Nano}^{form} = \frac{1}{N}(E_{Nano} - N_{Pb}\mu_{Pb} - N_{Se}\mu_{Se}) \quad (\ ) \quad 1$$

where E_{Nano}, μ_{Pb}, μ_{Se}, N_{Pb}, N_{Se} and N are the total energy of the system (nanostructure +matrix), the chemical potentials of Pb and Se, the number of atoms Pb and Se, and the total number of atoms ($N= N_{Pb}+N_{Se}$). Obviously, to calculate the formation energy, the chemical potentials of Pb and Se should be determined at the first step. The chemical potentials of constituent species can be varied to reflect specific equilibrium growth conditions[1]. Based on the ABF images (Figs. 2(d)-(g)) and the constructed model (Fig. 3(d)), containing the RS and ZB phases and additional Se atoms (the Se rich), it means that the ZB nanostructures is equilibrium with the PbSe-RS and Se compounds. Thus, from the Pb-Se phase diagram (Fig. S1(a)), μ_{Pb} and μ_{Se} should be constrained within the triangle region of RS-ZB-Se (the green region). And the chemical potentials can be obtained by solving the linear equations,

$$\begin{cases} \mu_{Pb} + \mu_{Se} = E_{PbSe}^{RS} \\ \mu_{Pb} + \mu_{Se} = E_{PbSe}^{ZB} \\ \mu_{Se} = E_{Se} \end{cases} \qquad (2)$$

where E_{PbSe}^{RS}, E_{PbSe}^{ZB}, and E_{Se} are the total energies of pristine bulk PbSe compounds in the RS and ZB phases and the Se compound, respectively. From DFT calculations, they are E_{PbSe}^{RS} = -8.22 eV/fu, E_{PbSe}^{ZB} = -7.84 eV/fu, and E_{Se} = -3.33 eV/fu. Unfortunately, it is unable to solve Eq 2. We then consider two limitations, the PbSe-RS and PbSe-ZB in equilibrium with Se, respectively (the red and blue lines in the left panel of Fig. S1). In these cases, we can get the chemical potentials of (μ_{Pb},μ_{Se}) are (-4.89, -3.33) eV/atom and (-4.51,-3.33) eV/atom at the RS-Se and ZB-Se equilibriums, respectively. Therefore, the corresponding formation energies of E_{Nano}^{form} are 91.4

meV/atom (RS-Se) and -104.5 meV/atom (ZB-Se). This means that the ZB nanostructures in RS matrix is thermodynamically unstable when the chemical potentials are extracting from the equilibrium between RS and Se. It is not surprising since there is no ZB phase in the matrix at all. However, once the chemical potentials are derived from the equilibrium between ZB and Se, the negative formation energy suggests the thermodynamical stability of the precipitated system. Since the chemical potential of Se is fixed at the Se-rich condition, we need to understand the stability behavior of the nanostructures precipitated system with varying the chemical potential of Pb (the right panel of Fig. S1). We notice that with increasing μ_{Pb} from μ^{RS} to μ^{ZB}, the sign of E_{Nano} changes from positive to negative, which means that the system undergoes thermodynamical unstabilization to stabilization. In other words, the nanostructures precipitated system is stable within the Pb chemical potential region of (-4.51, -4.71) eV/atom.

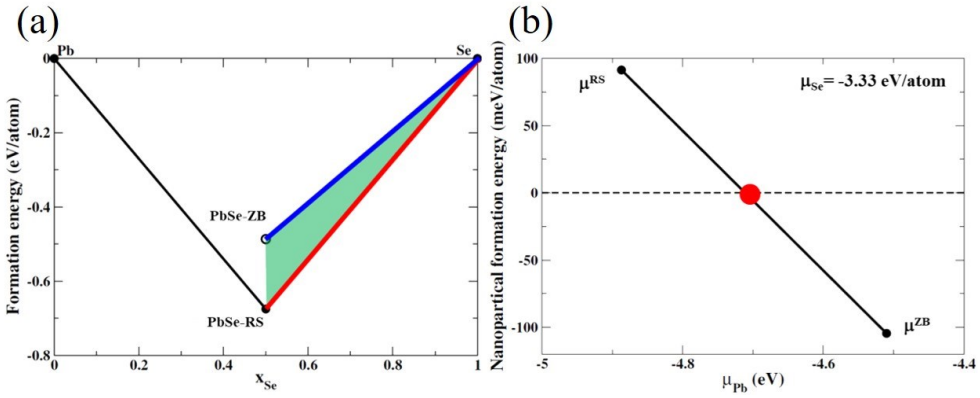

Fig. S1 (a) Theoretically determined Pb-Se phase diagram, (b) the formation energy of the nanostructures precipitated system with varying the chemical potential of Pb (μ_{Pb}) at μ_{Se}=3.33 eV/atom. The green region, red and blue lines represent the phase equilibriums of RS-ZB-Se, RS-Se and ZB-Se, respectively.

Supplementary figures and tables

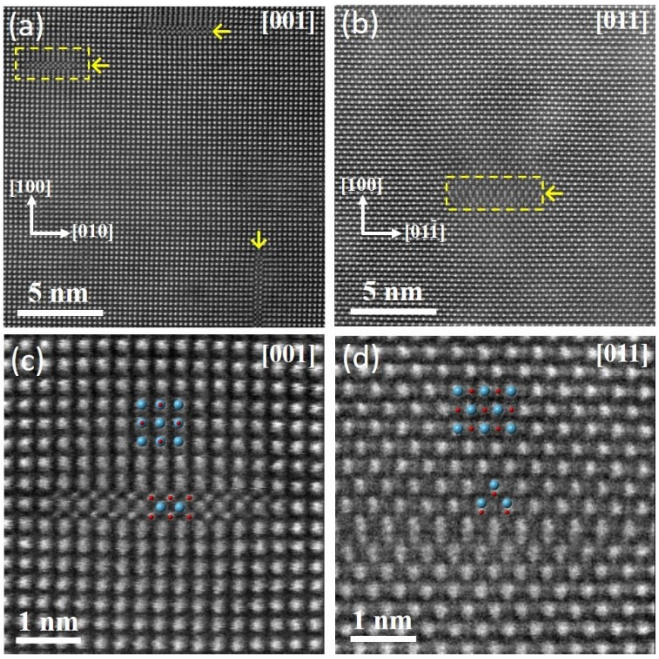

Fig. S2 The HAADF images corresponding to the Figure.2(d)-(g), respectively.

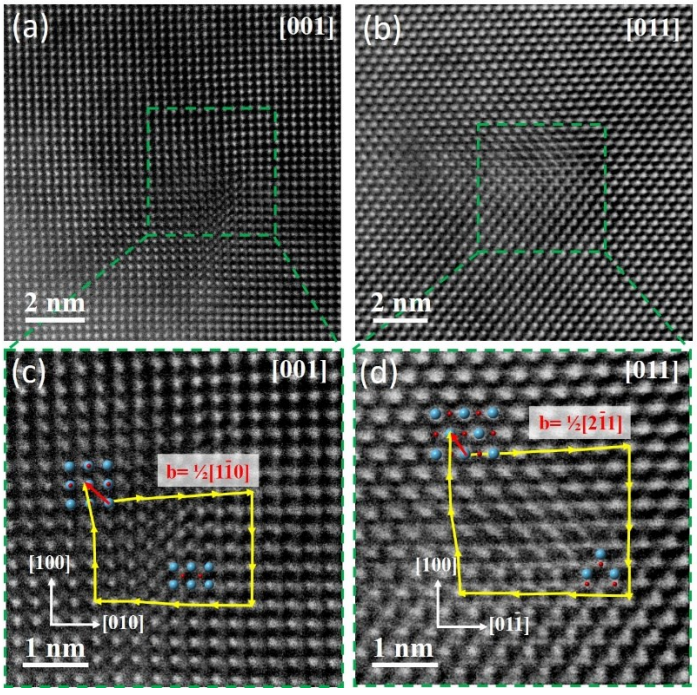

Fig. S3 The HAADF images corresponding to the Figure.4(a)-(d), respectively.

Table S1 The simulated structure of the zinc-blend (PbSe-ZB) phase precipitated in the rock-salt (PbSe-RS) matrix: a=62.060Å, b=24.824 Å and c=6.206 Å.

Atom	x	y	z
Pb1	0.7712	0.6936	0.9946
Pb2	0.7212	0.7111	0.4922
Pb3	0.6712	0.7111	0.9922
Pb4	0.6212	0.7111	0.4922
Pb5	0.5712	0.7111	0.9922
Pb6	0.5212	0.7111	0.4922
Pb7	0.4712	0.7111	0.9922
Pb8	0.4212	0.7111	0.4922
Pb9	0.3712	0.7111	0.9922
Pb10	0.3212	0.7111	0.4922
Pb11	0.2712	0.7111	0.9922
Pb12	0.7712	0.4085	0.9995
Pb13	0.7212	0.4085	0.4995
Pb14	0.6712	0.4085	0.9995
Pb15	0.6212	0.4085	0.4995
Pb16	0.5712	0.4085	0.9995
Pb17	0.5212	0.4085	0.4995
Pb18	0.4712	0.4085	0.9995
Pb19	0.4212	0.4085	0.4995
Pb20	0.3712	0.4261	0.9971
Pb21	0.3212	0.4261	0.4971
Pb22	0.2712	0.4261	0.9971
Pb23	0.3152	0.5483	0.024
Pb24	0.4192	0.5483	0.024
Pb25	0.5279	0.5442	0.0246
Pb26	0.6252	0.5401	0.0252
Pb27	0.7289	0.5442	0.0246
Pb28	0.2659	0.5526	0.5169
Pb29	0.3707	0.5526	0.5169
Pb30	0.4729	0.5526	0.5169
Pb31	0.5774	0.5485	0.5175
Pb32	0.6792	0.5444	0.518
Pb33	0.7782	0.5485	0.5175
Pb34	0.0213	0.8019	0.9981
Pb35	0.0713	0.9269	0.9981
Pb36	0.1213	0.8019	0.9981
Pb37	0.1713	0.9269	0.9981
Pb38	0.2213	0.8019	0.9981
Pb39	0.2713	0.9269	0.9981
Pb40	0.3213	0.8019	0.9981

Pb41	0.3713	0.9269	0.9981
Pb42	0.4213	0.8019	0.9981
Pb43	0.4713	0.9269	0.9981
Pb44	0.5213	0.8019	0.9981
Pb45	0.5713	0.9269	0.9981
Pb46	0.6213	0.8019	0.9981
Pb47	0.6713	0.9269	0.9981
Pb48	0.7213	0.8019	0.9981
Pb49	0.7713	0.9269	0.9981
Pb50	0.8213	0.8019	0.9981
Pb51	0.8713	0.9269	0.9981
Pb52	0.9213	0.8019	0.9981
Pb53	0.9713	0.9269	0.9981
Pb54	0.0213	0.5519	0.9981
Pb55	0.0713	0.6769	0.9981
Pb56	0.1213	0.5519	0.9981
Pb57	0.1713	0.6769	0.9981
Pb58	0.2213	0.5519	0.9981
Pb59	0.8213	0.5519	0.9981
Pb60	0.8713	0.6769	0.9981
Pb61	0.9213	0.5519	0.9981
Pb62	0.9713	0.6769	0.9981
Pb63	0.0213	0.3019	0.9981
Pb64	0.0713	0.4269	0.9981
Pb65	0.1213	0.3019	0.9981
Pb66	0.1713	0.4269	0.9981
Pb67	0.2213	0.3019	0.9981
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Pb73	0.8213	0.3019	0.9981
Pb74	0.8713	0.4269	0.9981
Pb75	0.9213	0.3019	0.9981
Pb76	0.9713	0.4269	0.9981
Pb77	0.0213	0.0519	0.9981
Pb78	0.0713	0.1769	0.9981
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Pb80	0.1713	0.1769	0.9981
Pb81	0.2213	0.0519	0.9981
Pb82	0.2713	0.1769	0.9981

Pb83	0.3213	0.0519	0.9981
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Pb87	0.5213	0.0519	0.9981
Pb88	0.5713	0.1769	0.9981
Pb89	0.6213	0.0519	0.9981
Pb90	0.6713	0.1769	0.9981
Pb91	0.7213	0.0519	0.9981
Pb92	0.7713	0.1769	0.9981
Pb93	0.8213	0.0519	0.9981
Pb94	0.8713	0.1769	0.9981
Pb95	0.9213	0.0519	0.9981
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Pb99	0.8713	0.0519	0.4981
Pb100	0.8213	0.1769	0.4981
Pb101	0.7713	0.0519	0.4981
Pb102	0.7213	0.1769	0.4981
Pb103	0.6713	0.0519	0.4981
Pb104	0.6213	0.1769	0.4981
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Pb146	0.7213	0.9269	0.4981
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Se170	0.1213	0.8019	0.4981
Se171	0.1213	0.9269	0.9981
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Se173	0.0213	0.9269	0.9981

Reference

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