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# Supporting Information

# Synergistic Defect Engineering for Improving n-type NbFeSb Thermoelectric Performance through High-Throughput Computations

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Table S1. The chemical potentials (in eV) of NbFeSb at the A, B, C, D, E and F points labeled in Fig. 2.

| Point | Fe | Sb | Nb |
|---|---|---|---|
| A | -8.71 | -4.35 | -10.597 |
| B | -8.68 | -4.48 | -10.497 |
| C | -8.37 | -4.96 | -10.327 |
| D | -7.80 | -4.38 | -11.447 |
| E | -8.43 | -4.08 | -11.147 |
| F | -8.25 | -4.16 | -11.247 |

Table S2: Chemical potentials of extrinsic elements after considering all possible phases from the OQMD database.

| Elements | Chemical potentials | considered second phases |
|---|---|---|
| Y | -8.568 | $Y_5Sb_3$/$NbSb_2$/$NbFe_2$/$Nb_5Sb_4$/$YFe_5$/$Nb_3Sb$/$YFe_2$//YSb/$FeSb_3$/$FeSb_2$ |
| Hf | -10.716 | $NbFe_2$/$FeSb_3$/$FeSb_2$/$Nb_3Sb$/$NbSb_2$/$HfFe_2$/HfFe/$Hf_5Sb_3$/HfSb/$HfSb_2$/$Hf_6FeSb_2$/$Nb_5Sb_4$/ |
| Zr | -9.779 | $Nb_5Sb_4$/$FeSb_2$/$Zr_5Sb_3$/ZrNbSb/$Zr_6FeSb_2$/$NbSb_2$/$FeSb_3$/ZrSb/$NbFe_2$/$Nb_3Sb$/$ZrSb_2$/$Zr_3Fe$/$Zr_2Sb$/$ZrFe_2$ |
| Sc | -7.97 | ScSb/$NbSb_2$/$NbFe_2$/$Nb_5Sb_4$/$ScFe_2$/$Nb_3Sb$/$Sc_2Sb$/$Sc_5Sb_3$/$Sc_6FeSb_2$/$FeSb_3$/ $FeSb_2$/ |
| Ta | -12.128 | $TaNbSb_4$/$NbSb_2$/$NbFe_2$/$TaSb_2$/$Ta_5Sb_4$/$Ta_2Nb$/$Nb_3Sb$/$TaNb_4Sb_4$/$TaFe_2$/$Ta_3Fe$/TaFe/$FeSb_3$/TaFeSb/$FeSb_2$ |
| W | -12.87 | $Nb_3Sb$/$Fe_2W$/$NbW_3$/$NbSb_2$/$Nb_5Sb_4$/$Fe_7W_6$/$FeSb_3$/$NbFe_2$/$FeSb_2$/$FeSb_2$ |
| Ti | -8.498 | $NbSb_2$/TiSb/$Ti_3Nb_3Sb_2$/TiFe/TiNb/$TiFe_2$/$NbFe_2$/$Nb_3Sb$/$Nb_5Sb_4$/$FeSb_2$/$FeSb_3$/$TiSb_2$/$Ti_2Fe$/$Ti_5Sb_3$/$Ti_3Sb$/$Ti_2Fe_3Sb_2$/$Ti_2Sb$ |
| Mo | -10.635 | $Nb_5Sb_4$/$FeSb_2$/NbMo/$NbSb_2$/$FeSb_3$/$Nb_3Mo_{13}$/$Nb_9Mo_7$/$NbFe_2$/$Nb_3Sb$/$Fe_2Mo$/$NbMo_3$/$Sb_7Mo_3$ |
| Re | -12.377 | $Fe_2Re_3$/NbRe/$FeSb_3$/$NbFe_2$/$Nb_2Re$/$FeSb_2$/$Nb_3Re$/$Nb_3Sb$/$NbSb_2$/$Nb_5Sb_4$/$Nb_{11}Re_5$/$Fe_3Re$/FeRe/$Nb_5Re_{24}$/ |
| V | -9.147 | VFeSb/$V_3Sb$/$Nb_2VFe_3$/$Nb_2V_3Fe$/$FeSb_3$/$NbFe_2$/$FeSb_2$/$Nb_3Sb$/$NbSb_2$/$Nb_5Sb_4$/$V_3Fe$/$VFe_3$ |
| Os | -11.297 | $Nb_3Sb$/$FeSb_2$/$NbFe_2$/$Sb_2Os$/$Nb_3Os$/$Nb_2Fe_3Os$/$FeSb_3$/NbSbOs/$NbSb_2$/$Nb_5Sb_4$ |
| Cr | -7.47 | $FeSb_3$/$Nb_3Sb$/$NbSb_2$/$FeSb_2$/$Nb_5Sb_4$/$NbCr_2$ |
| Ru | -9.716 | $Nb_5Sb_4$/$FeSb_2$/NbSbRu/$NbSb_2$/$NbRu_3$/$FeSb_3$/$Nb_3Ru$/$Sb_2Ru$/$NbFe_2$/$Nb_3Sb$/NbRu |
| Ir | -9.375 | $NbIr_3$/$NbSb_2$/$Sb_3Ir$/$NbFe_2$/$Nb_5Sb_4$/$Nb_3Sb$/$Fe_3Ir$/$Nb_2Fe_3Ir$/$Nb_3Ir$/$Sb_2Ir$/$Nb_4Sb_5Ir_5$/$FeIr_3$/$FeSb_3$/$FeSb_2$/NbIr |
| Mn | -8.828 | MnSb/$Nb_3Sb$/$FeSb_2$/$NbFe_2$/$MnNb_2Fe_3$/MnNb/$Mn_2Nb$/$FeSb_3$/ |

| | | |
|---|---|---|
| | | $Mn_3Nb_2Fe/NbSb_2/Nb_5Sb_4$ |
| Rh | -8.05 | $Nb_5Sb_4/SbRh/FeSb_2/NbRh/FeRh_3/Fe_2Rh/NbSb_2/FeSb_3/NbFe_2$ /$Nb_3Sb/NbRh_3/Sb_3Rh/Nb_4Sb_5Rh_5/Sb_2Rh/Nb_3Rh/SbRh_2$ |
| Pt | -6.144 | $Nb_5Sb_4/FeSb_2/Nb_3Pt/NbPt_8/FePt_5/NbSb_2/Sb_2Pt/FeSb_3/NbPt/S$ $b_2Pt_3/Fe_3Pt_5/NbFe_2/Nb_3Sb/FePt_3/SbPt/NbPt_2/NbPt_3/FePt/$ |
| Pd | -5.836 | $Nb_5Sb_4/Sb_2Pd/FeSb_2/NbPd_5/NbSb_2/NbPd_2/NbPd_3/Sb_7Pd_{20}/Fe$ $Pd_5/FeSb_3/FePd_3/NbPd_8/FePd_2/NbFe_2/Nb_3Sb/SbPd_5/SbPd/Sb$ $Pd_2/Sb_8Pd_{11}$ |
| Co | -7.398 | $CoSb_3/FeCo/CoSb/FeSb_3/Fe_9Co_7/NbFe_2/FeSb_2/FeSb_2/NbCo_3/$ $Fe_3Co/Nb_3Sb/Fe_7Co/NbSb_2/Nb_7Co_6/Nb_5Sb_4/Fe_{11}Co_5/Fe_{13}Co_3/$ $Nb_4Co_5Sb_5/Fe_5Co_3/Nb_2Co$ |
| Au | -2.23 | $NbAu_2/NbSb_2/FeSb_2/FeSb_3/Nb_3Sb/Sb_2Au/Nb_3Au_2/NbFe_2/Nb_5$ $Sb_4$ |
| Ni | -5.332 | $FeNi_3/FeSb_2/Nb_3Ni_5Sb_5/Ni_5Sb_2/FeSb_3/Nb_3Sb/NiSb/FeNiSb_4/$ $Ni_3Sb/FeNi/$ $NbNi_3/NbSb_2/NiSb_2/NbFe_2/FeNi_8/Nb_5Sb_4/Nb_7Ni_6$ |
| Ag | -0.83 | $Nb_3Sb/NbSb_2/Nb_5Sb_4/FeSb_3/NbFe_2/FeSb_2$ |
| Cu | -1.97 | $NbSb_2/Nb_5Sb_4/FeSb_3/NbFe_2/Nb_3Sb/FeSb_2$ |
| Zn | -2.068 | $Nb_6Zn_7/NbSb_2/ZnSb_2/NbZnFe_4/NbFe_2/Nb_3Sb/Nb_5Sb_4/Zn_{13}Fe$ /$FeSb_3/FeSb_2/NbZn_3/NbZn_2/ZnSb/Zn_3Fe$ |
| Cd | -0.56 | $NbSb_2/CdSb/FeSb_2/Nb_3Sb/FeSb_3/NbFe_2/Nb_5Sb_4$ |
| As | -5.5 | $FeAs_2/SbAs/NbAs_2/Nb_3Sb/FeSb_2/NbFe_2/Nb_5As_3/FeAs/FeSb_3/$ $NbAs/Nb_3As/NbSb_2/Nb_5Sb_4/NbFeAs$ |
| Bi | -4.39 | $Nb_3Sb/BiSb/NbSb_2/Nb_5Sb_4/FeSb_3/NbFe_2/FeSb_2/$ |
| Ge | -4.29 | $Nb_5Sb_4/Nb_2Fe_3Ge/FeSb_2/NbSb_2/NbGe_2/FeSb_3/FeGe/NbFe_2/N$ $b_3Sb/Fe_3Ge/NbFeGe/Nb_5Ge_3/NbGeSb$ |
| Pb | -3.951 | $NbSb_2/Nb_5Sb_4/FeSb_3/NbFe_2/Nb_3Sb/FeSb_2$ |
| Sn | -4 | $NbSb/NbSb_2/NbFe_4Sn/NbFe_2/Nb_3Sb/Nb_5Sb_4/FeSb_2/FeSb_3/Fe$ $Sn_2/Sn_4Sb_3/NbSn_2/Nb_3Sn/Nb_6SnSb/FeSn$ |
| Al | -4 | $NbSb_2/NbFe_2/Nb_5Sb_4/Nb_3Sb/NbAl_3/AlFe/NbAlFe_2/AlFe_3/Al_6$ $Fe/Nb_2Al/Al_9Fe_2/FeSb_3/Al_2Fe/AlSb/FeSb_2$ |
| Ga | -3.754 | $NbSb_2/NbFe_2/Nb_3Sb/Nb_5Sb_4/Nb_5Ga_{13}/FeSb_3/GaFe_3/FeSb_2/Nb$ $_3Ga_2/NbGa_3/GaSb/Ga_3Fe/NbGaFe_2$ |
| In | -2.72 | $NbSb_2/InSb/FeSb_3/NbFe_2/Nb_3Sb/Nb_5Sb_4/FeSb_2$ |
| Tl | -2.48 | $Nb_3Sb/NbSb_2/Nb_5Sb_4/FeSb_3/Tl_7Sb_2/NbFe_2/FeSb_2/$ |
| Si | -6.502 | $NbSb_2/Nb_5Si_3/Nb_6Fe_{16}Si_7/NbFe_2/Nb_3Sb/FeSi/Nb_5Sb_4/FeSb_2/F$ $eSb_3/NbFeSi/Nb_2Fe_3Si/Fe_3Si/NbSi_2/FeSi_2/NbSiSb$ |
| P | -7.147 | $FeP_4/Fe_2P/Nb_3P/FeP_2/Nb_3Sb/FeSb_2/NbFeP/NbFe_2/Nb_2P/FeSb$ $_3/NbP/NbSb_2/Fe_3P/SbP_5/Nb_5Sb_4/Nb_7P_4/FeP$ |
| S | -4.738 | $NbS_2/NbS_3/FeS/NbS_5/Nb_3FeS_6/FeSb_3/Nb_{21}S_8/NbFe_2/FeSb_2/S$ $bS_2/Nb_3Sb/Nb_3S_4/Sb_2S_3/NbSb_2/Nb_5Sb_4/FeSbS/FeS_2$ |
| Se | -3.644 | $NbSb_2/NbFe_2/Nb_5Sb_4/Nb_3Sb/NbSe_2/FeSe_2/Nb_2Se/Sb_2Se_3/Nb$ $Se_3/FeSbSe/Nb_3Se_4/FeSb_3/FeSb_2/Nb_2Se_9$ |
| Te | -3.25 | $NbTe_4/NbSb_2/Nb_3Te_4/NbFe_2/FeTe_2/Nb_3Sb_2Te_5/Nb_5Sb_4/Nb_3S$ |

| | | |
|---|---|---|
| | | b/$Fe_2Sb_3Te$/$NbTe_2$/$Nb_3Te_2$/$Sb_2Te_3$/$FeSb_2$/$FeSb_3$ |
| B | -7.408 | Nb3B4/Nb2Sb/FeSb2/NbFe2/Fe2B/NbB2/Nb3B2/Nb2B3/FeSb3/NbB/Nb5B6/NbSb2/NbFeB/FeB/Nb5Sb4/NbFeB3 |
| C | -9.044 | $Nb_5Sb_4$/$FeSb_3$/$Nb_2C$/$Nb_3Sb$/$NbSb_2$/$FeSb_2$/$NbFe_2$/$Nb_6C_5$ |
| N | -8.184 | $Nb_5N_6$/$Nb_2N$/$FeSb_3$/$NbFe_2$/$FeSb_2$/FeN/$Nb_3Sb$/$NbSb_2$/NbN/$Nb_5Sb_4$/$Fe_3N$ |
| O | -8.331 | NbO/$Nb_5Sb_4$/$FeSb_2$/$FeSb_2O_6$/$Nb_2FeO_6$/FeO/$NbO_2$/$NbSb_2$/$FeSb_2O_4$/$FeSb_3$/$Sb_2O_3$/$Fe_2O_3$/$Nb_2O_5$/$NbFe_2$/$Nb_3Sb$/$NbFeO_4$/$SbO_2$/$NbSbO_4$/$Fe_3O_4$/$Sb_2O_5$ |
| F | -1.578 | $SbF_5$/$FeF_3$/$Nb_3Sb$/$FeF_4$/$FeSb_2$/$NbFe_2$/$NbF_5$/$Nb_2F_5$/$SbF_3$/$FeF_2$/$FeF_6$/$Sb_2F_7$/$FeSb_3$/$NbFeF_6$/$NbSb_2$/$Nb_5Sb_4$ |
| Cl | -0.379 | $Nb_3Cl_8$/$SbCl_5$/$FeSb_3$/$NbFe_2$/$NbCl_4$/$FeCl_2$/$FeSb_2$/$FeCl_4$/$Nb_3Sb$/$SbCl_{11}$/$NbCl_5$/$NbSb_2$/$Nb_5Sb_4$/$Nb_3Cl_7$/$FeCl_3$/$SbCl_3$ |
| Br | -0.472 | $NbBr_4$/$Nb_3Br_8$/$FeSb_3$/$FeBr_2$/$NbBr_5$/$NbFe_2$/$SbBr_7$/$FeSb_2$/$FeBr_3$/$Nb_3Sb$/$NbSb_2$/$Nb_5Sb_4$/$SbBr_3$/$Fe_3Br_8$ |
| I | -0.23 | $Nb_5Sb_4$/$FeSb_2$/$NbI_3$/$SbI_3$/$NbSb_2$/$FeI_2$/$FeI_3$/$NbSb_2$/$FeI_2$/$FeSb_3$/$NbFe_2$/$NbI_5$/$Nb_3Sb$/$NbI_4$ |

Table S3. The Debye temperature Θ (K), Grüneisen parameter γ, phonon velocities $v$ (m/s) of transverse (TA/ TA') and longitudinal (LA) acoustic branches in NbFeSb.

| | LA | TA | TA' |
|---|---|---|---|
| Θ | 194 | 196 | 259 |
| γ | 1.68 | 1.55 | 1.86 |
| $\mathcal{V}$ | 2866 | 3227 | 5851 |

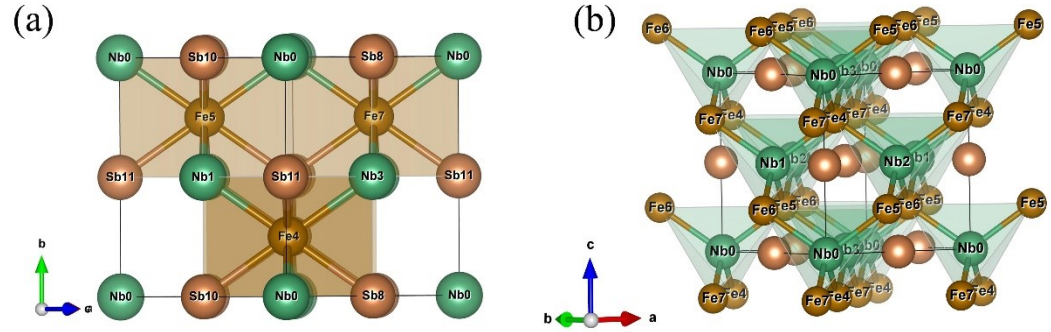



Figure S1. The illustration local geometry of atoms in NbFeSb. (a) The Fe atoms are with the hexahedron environment, while (b) Nb atoms at the tetrahedral sites.

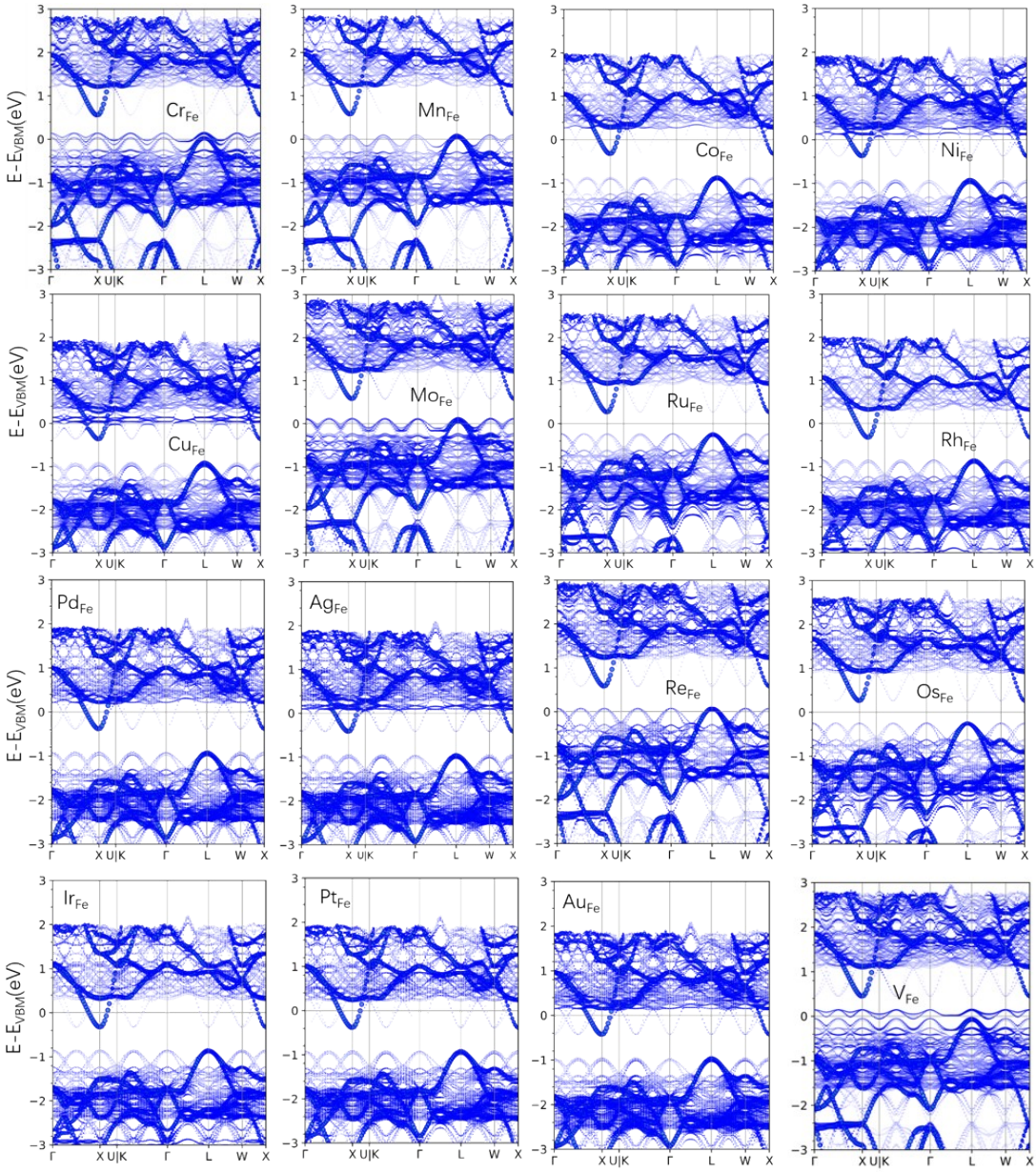
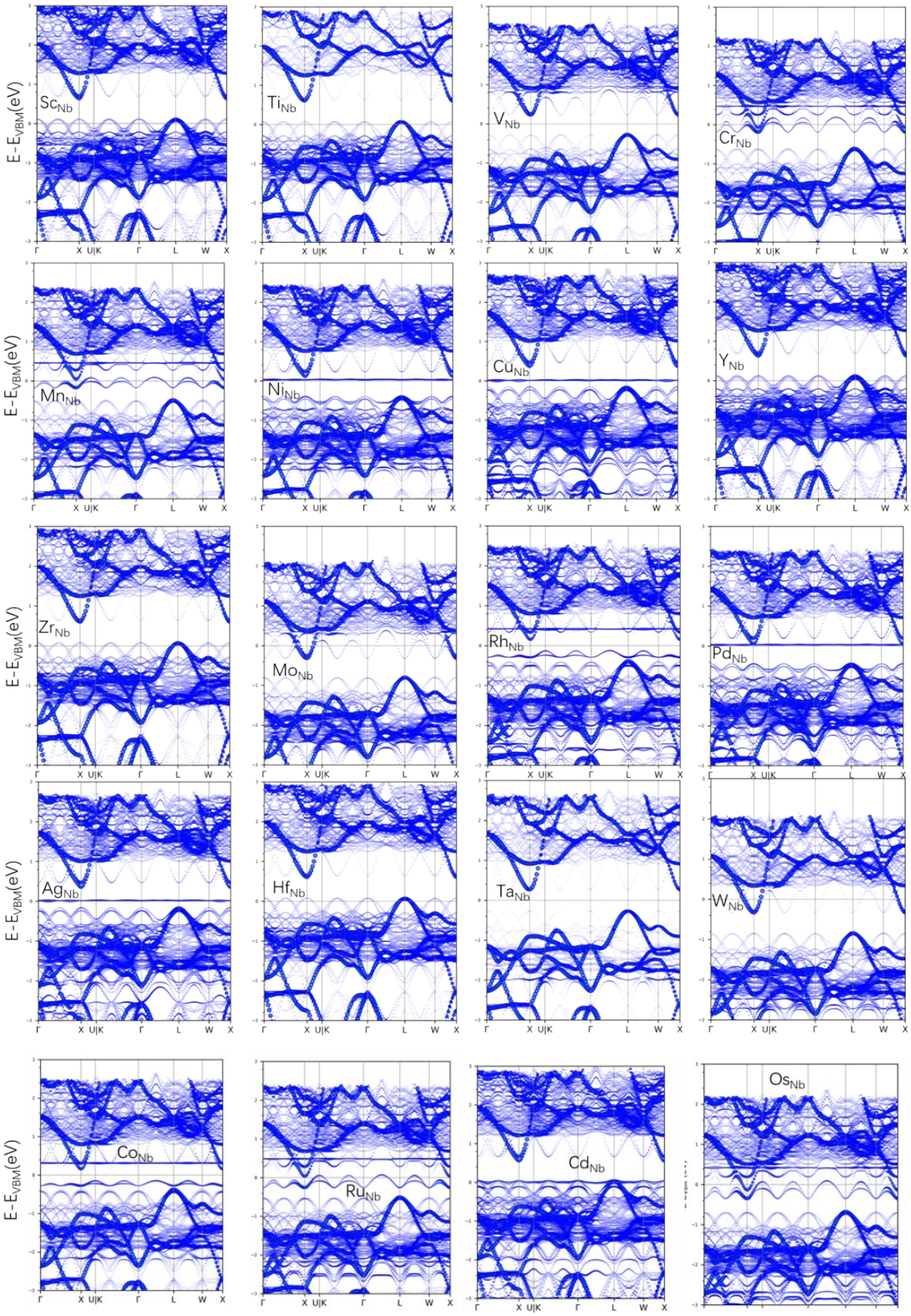



Figure S2. The unfolding band structures of $NbFe_{0.97}X_{0.03}Sb$ (X=Cr/Mn/Co/Ni/Cu/Mo/Ru/Rh/Pd/Ag/Re/Os/Ir/Pt/Au/V) compounds.


E-E$_{VBM}$(eV)
Sc$_{Nb}$
Ti$_{Nb}$
V$_{Nb}$
Cr$_{Nb}$
Mn$_{Nb}$
Ni$_{Nb}$
Cu$_{Nb}$
Y$_{Nb}$
Zr$_{Nb}$
Mo$_{Nb}$
Rh$_{Nb}$
Pd$_{Nb}$
Ag$_{Nb}$
Hf$_{Nb}$
Ta$_{Nb}$
W$_{Nb}$
Co$_{Nb}$
Ru$_{Nb}$
Cd$_{Nb}$
Os$_{Nb}$
Γ X U|K Γ L W X

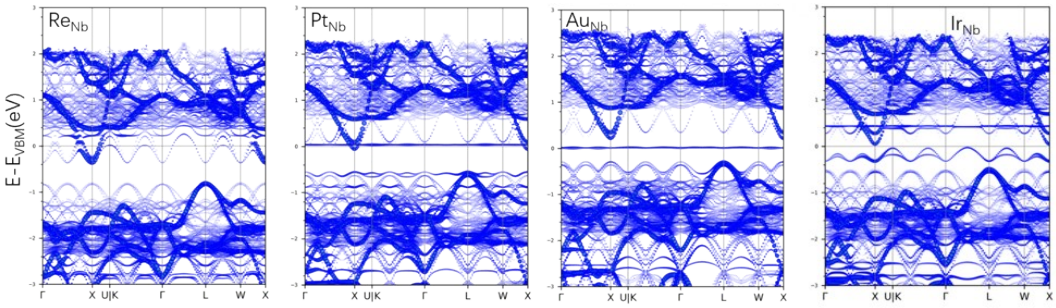



Figure S3. The unfolding band structures of $Nb_{0.97}X_{0.03}FeSb$ (X=Sc/Ti/V/Cr/Mn/Ni/Cu/Y/Zr/Mo/Rh/Pd/Ag/Hf/Ta/W/Co/Ru/Cd/Os/Re/Pt/Au/Ir) compounds.

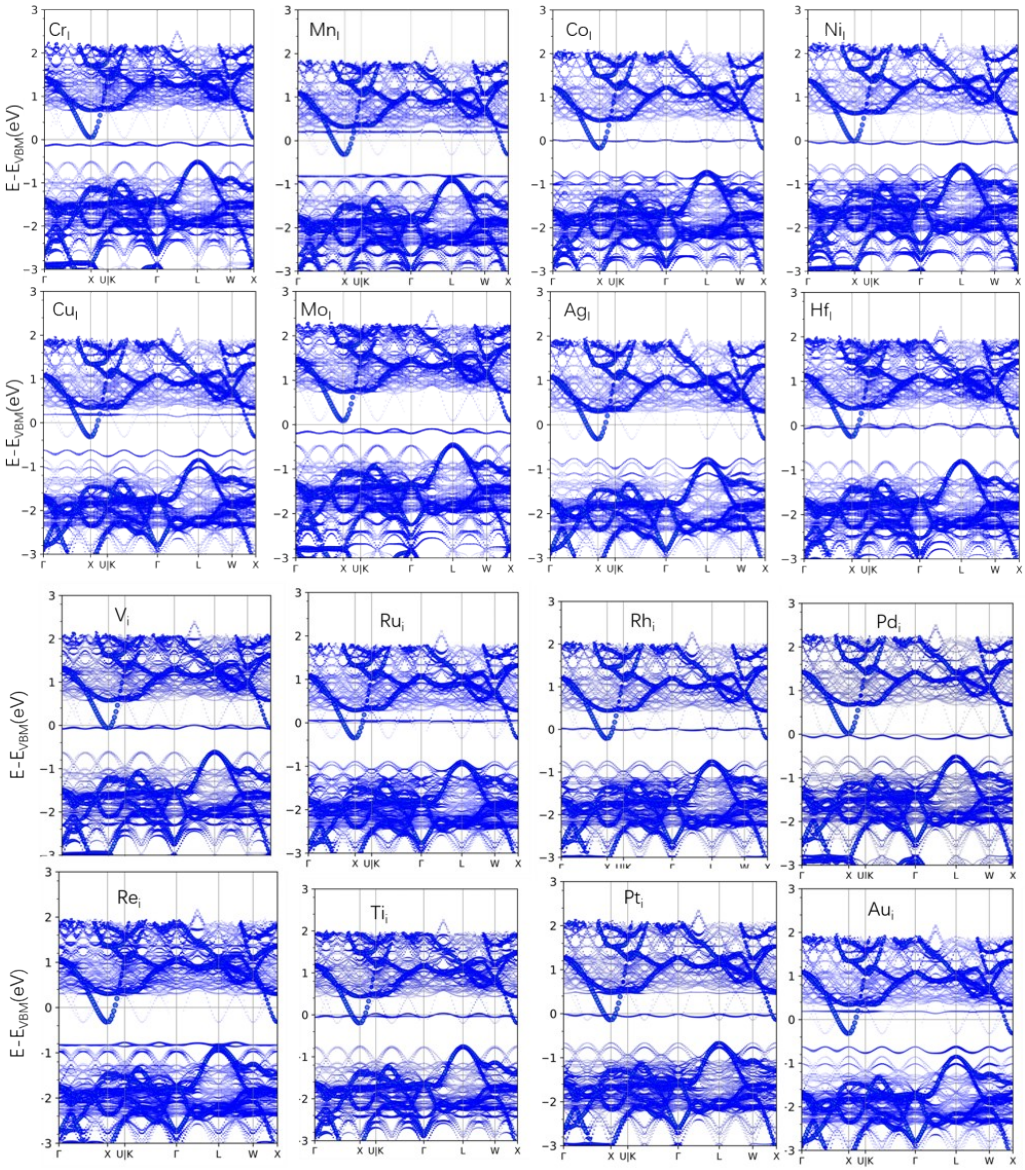



Figure S4. The unfolding band structures of $NbFeX_{0.03}Sb$ (X=Cr/Mn/Co/Ni/Cu/Mo/Ag/Hf/V/Ru/Rh/Pd/Re/Ti/Pt/Au) compounds.

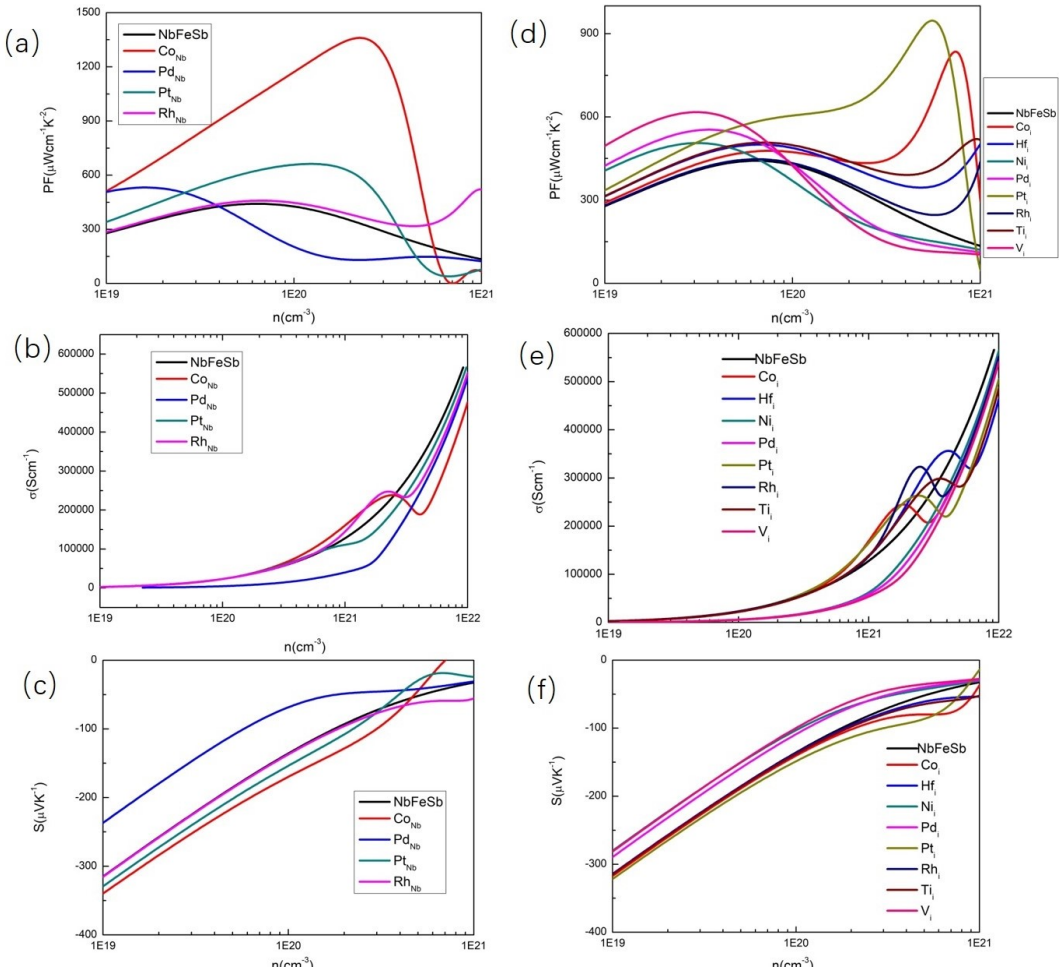



Fig. S5. (a) The power factor, (b) electrical conductivity, and (c) Seebeck coefficient of $Co_{Nb}$, $Pd_{Nb}$, $Pt_{Nb}$, $Rh_{Nb}$ defects in NbFeSb material. (e) The power factor, (d) electrical conductivity, and (f) Seebeck coefficient of $Pt_i$, $Co_i$, $Hf_i$, $Ni_i$, $Pd_i$, $Rh_i$, $Ti_i$, $V_i$ defects in NbFeSb materials.