On the origin of in-gap states in amorphous $Ge_2Sb_2Te_5$ Supplementary Material

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Contents

1 Electronic DOS of NN models

In Figs. S1-S30 we show the electronic density of states (DOS) computed with the HSE hybrid functional [1] of the thirty 297-atom amorphous models of GST generated by quenching the melt in NN-MD and then geometry-optimized at zero temperature with the HSE functional. Isosurfaces of the most localized in-gap states analyzed in the article are also shown (see Table S2).

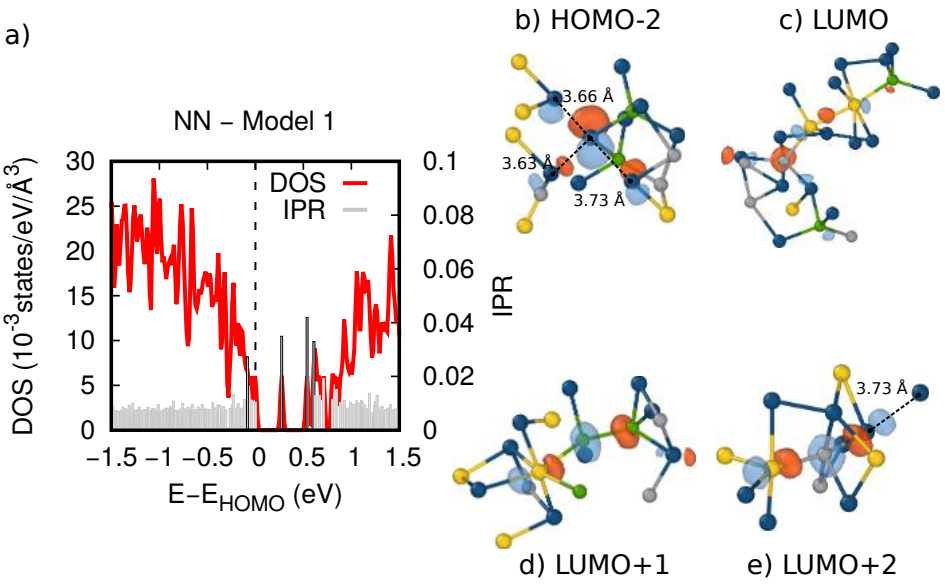

Figure S1: a) A zoom of the electronic DOS of model NN-1 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b)-e). b) The HOMO-2 is localized on a 2-coordinated Te atom bonded to two Ge_T atoms and forming there long Te-Te contacts (3.63- 3.73 Å). c) The LUMO is localized close to a 5-coordinated Ge atom with one wrong bond with a partial contribution of a small chain of four wrong bonds (Ge-Sb-Sb-Ge_T). d) The LUMO+1 is localized in the neighborhood of two bonded Ge_T atoms and an overcoordinated Sb atom. e) LUMO+2 is localized near a 5-coordinated Ge atom with two wrong bonds. In all panels, Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

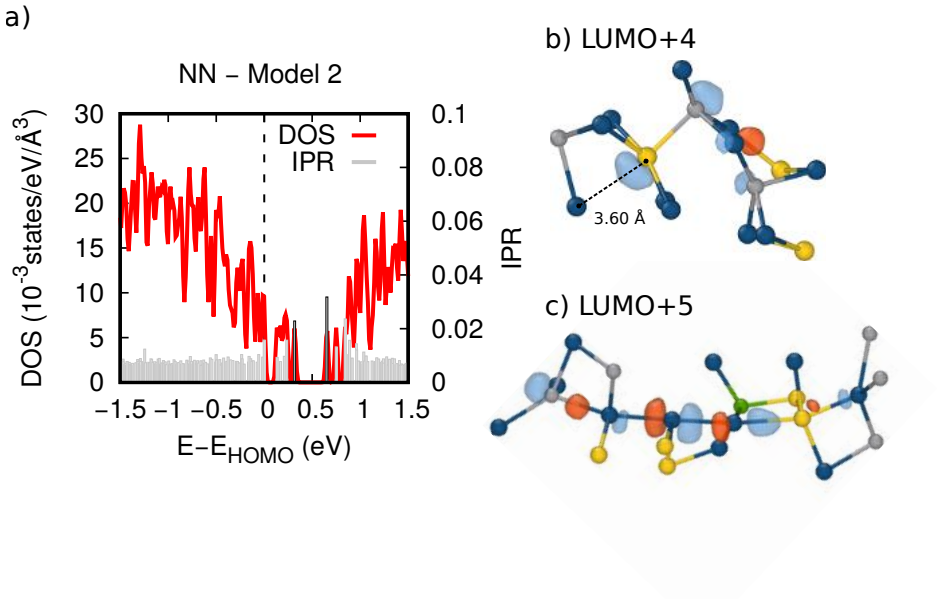

Figure S2: a) A zoom of the electronic DOS of model NN-2 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO+4 is localized in the neighborhood of a wrong bond between a 5-coordinated Sb and a 4-coordinated Ge atom. The Sb atom also forms a long contact with a Te atom (3.60 Å). c) The LUMO+5 is localized on a chain of Te atoms. In all panels, Ge, Ge_{Td} (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

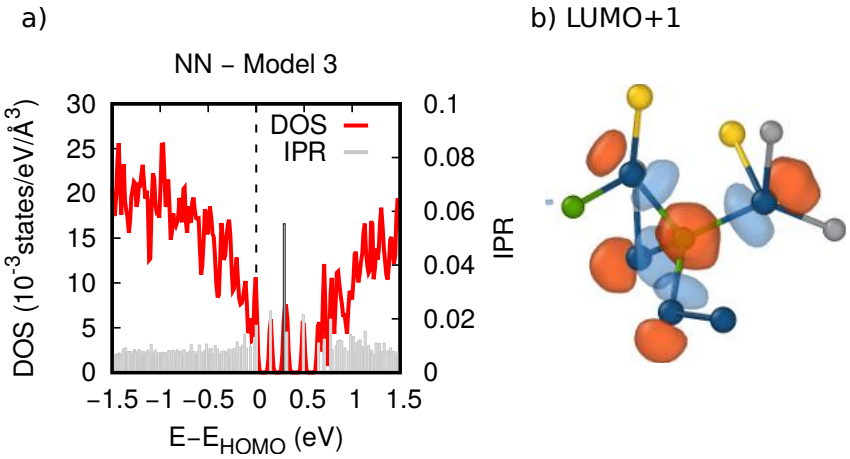

Figure S3: a) A zoom of the electronic DOS of model NN-3 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO+1 is localized on a Ge_T bonded to four Te atoms. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

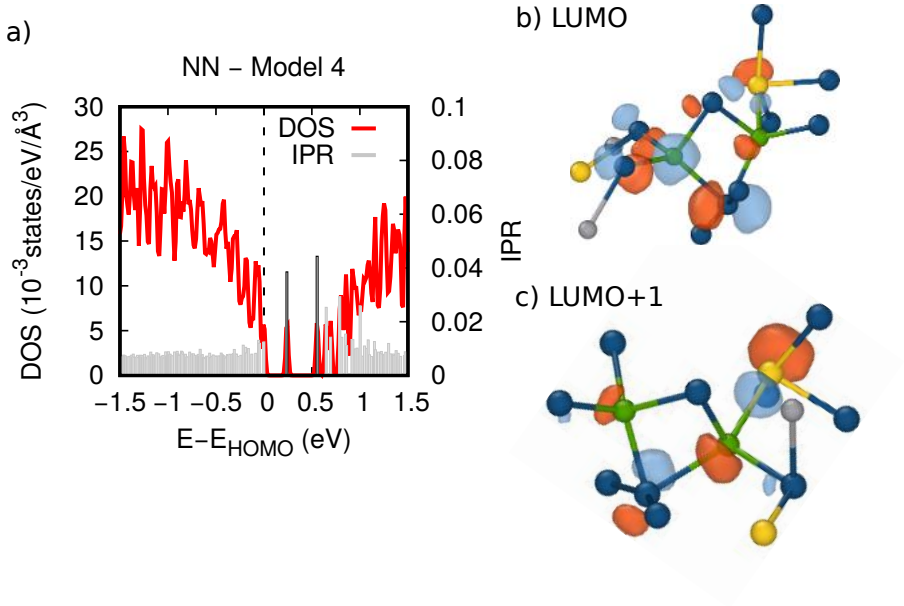

Figure S4: a) A zoom of the electronic DOS of model NN-4 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized close to a couple of Ge_T atoms bonded mainly to Te atoms and one wrong Ge-Sb bond. c) The LUMO+1 is localized on the same environment of the LUMO state. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

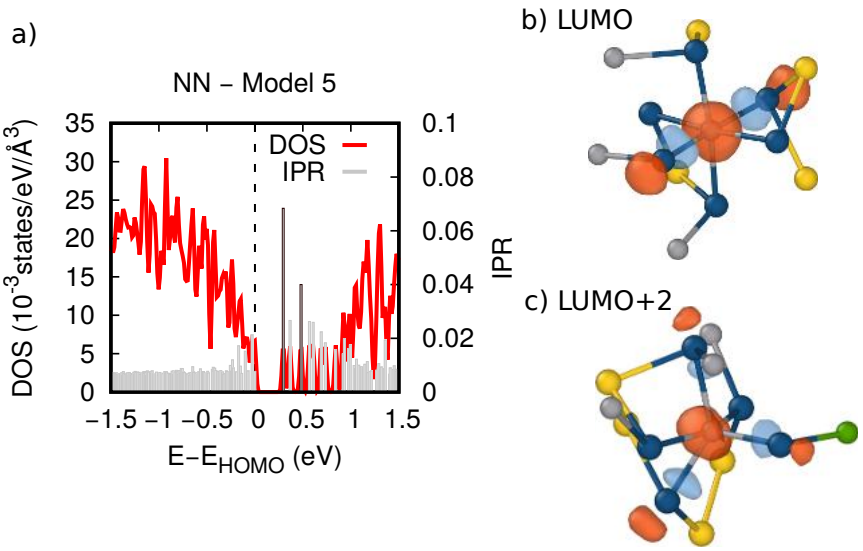

Figure S5: a) A zoom of the electronic DOS of model NN-5 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. The LUMO (panel b) and the LUMO+2 (panel c) are localized on overcoordinated Ge atoms. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

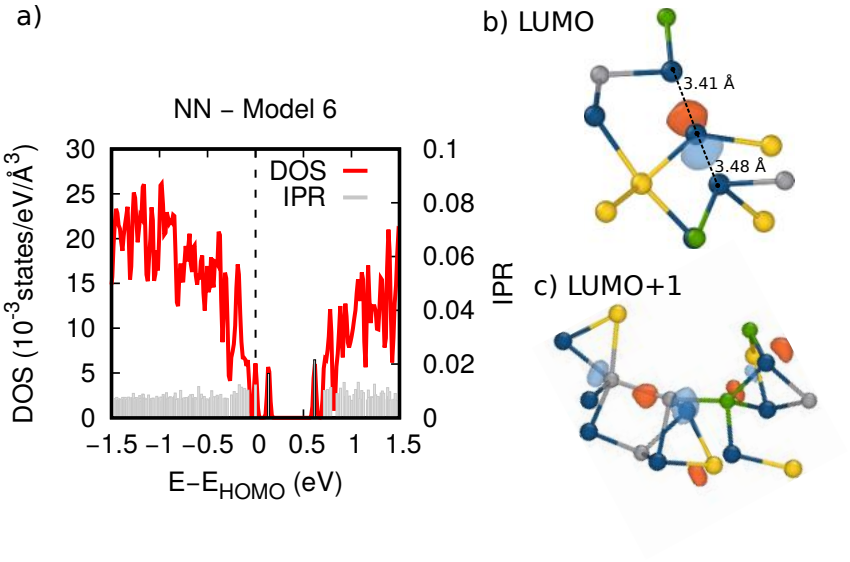

Figure S6: a) A zoom of the electronic DOS of model NN-6 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is weakly localized on a 2-coordinated Te atoms forming two long bonds with Te atoms. c) The LUMO+1 is localized on a long chain of Ge atoms involving overcoordinated and tetrahedral atoms. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

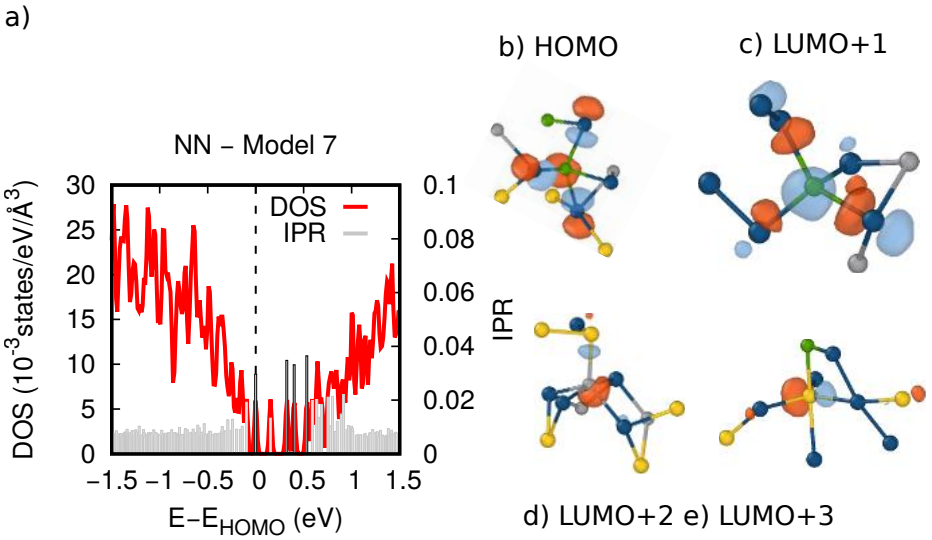

Figure S7: a) A zoom of the electronic DOS of model NN-7 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b)-e). b) The HOMO state which is localized on a 4-coordinated Ge atom with a q-order parameter of 0.78 very close to that of a Ge_T atom. c) The LUMO+1 is localized on a Ge_T atom. d) The LUMO+2 is localized on a 5-coordinated Ge atom with a wrong bond. e) LUMO+3 is localized on a 5-coordinated Sb atom bonded to a Ge_T atom. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

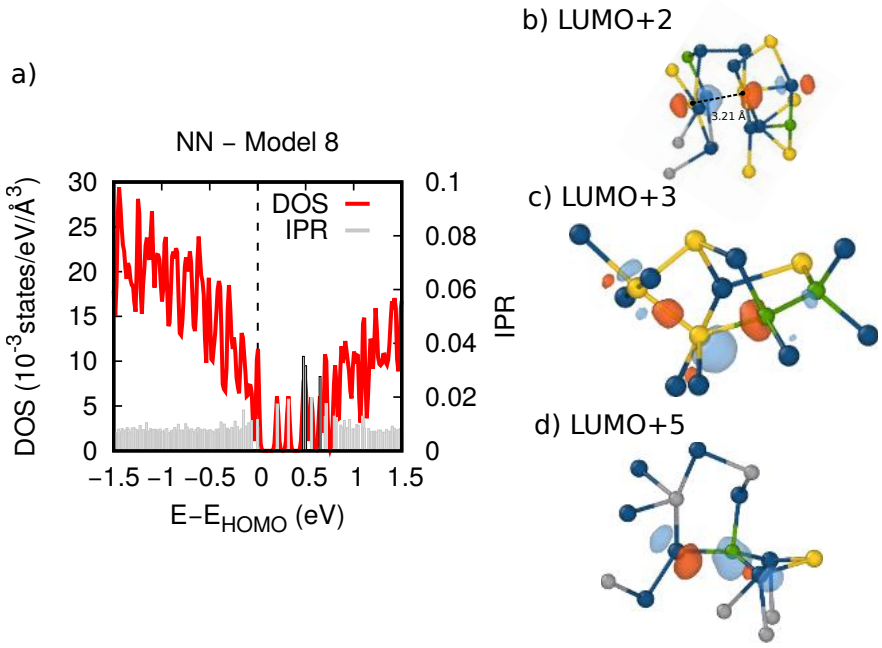

Figure S8: a) A zoom of the electronic DOS of model NN-8 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b)-d). b) The LUMO+2 is localized on two Sb atoms forming a bond slightly longer than the cutoff. c) The LUMO+3 is localized on a chain of wrong Sb-Sb and Sb-Ge_T wrong bonds. d) The LUMO+5 is on a Ge_T atom. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

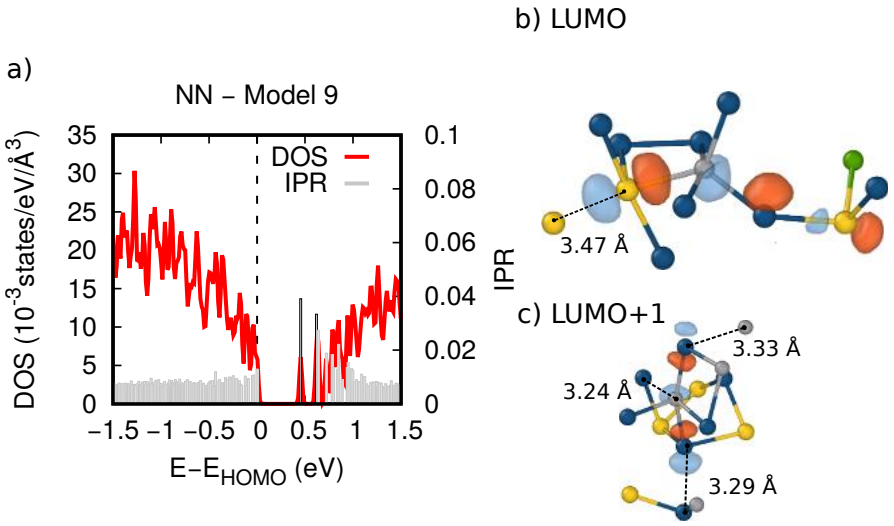

Figure S9: a) A zoom of the electronic DOS of model NN-9 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, and c. b) The LUMO is localized close to an overcoordinated Ge atom with a Sb-Ge wrong bond and on an Sb atom forming a bond slightly longer than the cutoff. c) The LUMO+1 is localized on an a Ge-atom in a defective octahedral geometry. Ge, Ge_{γ} (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

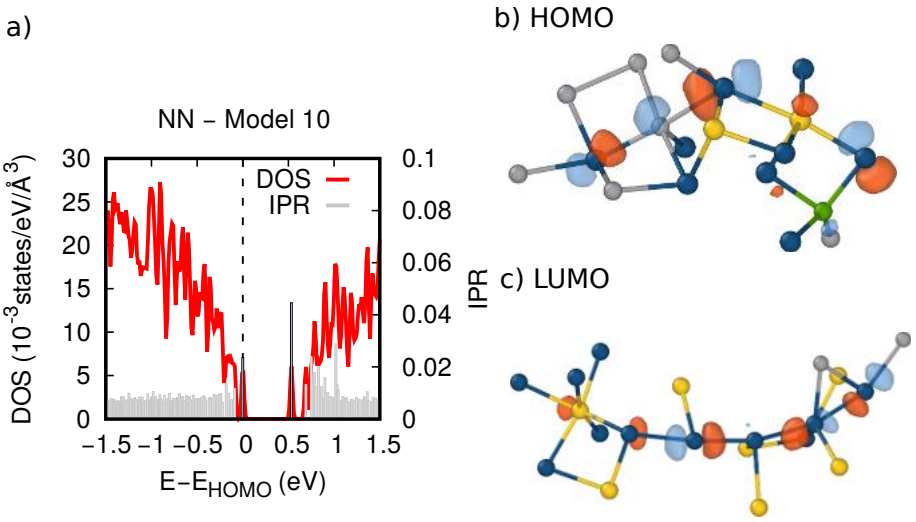

Figure S10: a) A zoom of the electronic DOS of model NN-10 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, and c. b) The HOMO is localized on the neighborhood of an overcoordinated Ge and Sb atoms in a defective octahedral geometry. c) The LUMO is localized on a long chain of Te-Te bonds. Ge, Ge_T (tetrahedral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

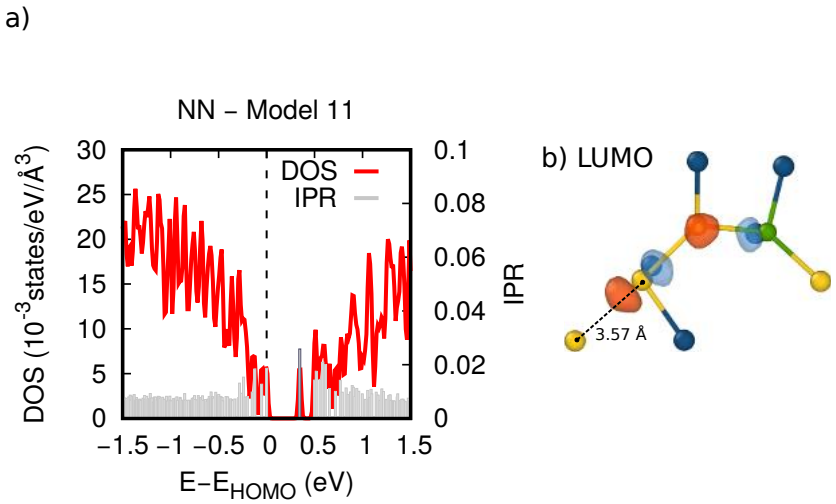

Figure S11: a) A zoom of the electronic DOS of model NN-11 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO is localized on a chain of Sb-Sb-Ge_{γ}-Sb wrong bonds. Ge, Ge_{γ} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

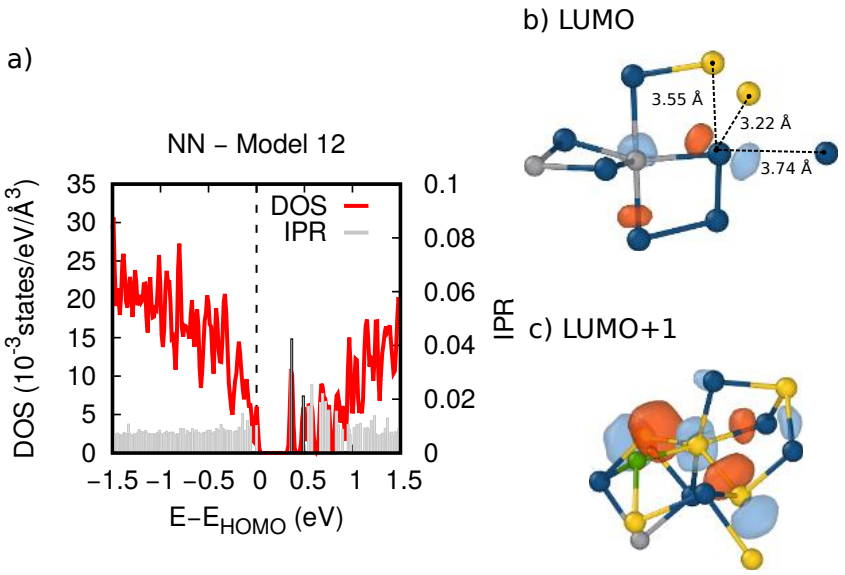

Figure S12: a) A zoom of the electronic DOS of model NN-12 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, and c. b) The LUMO is localized on a 5-coordinated Ge atom bonded to a 2-coordinated Te atom. The latter forms three long contacts with Sb and Te atoms. c) The LUMO+1 is localized on an overcoordinated Sb atom with several wrong bonds. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

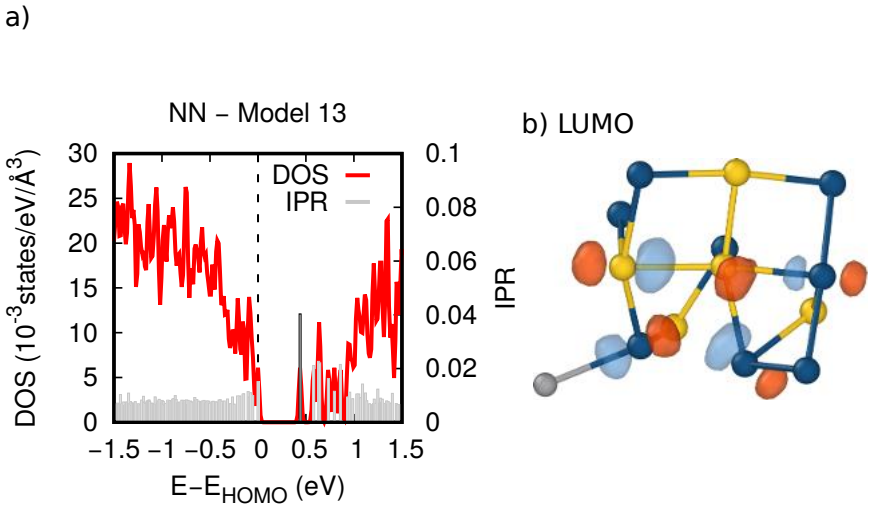

Figure S13: a) A zoom of the electronic DOS of model NN-13 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO is localized close to a 5-coordinated Sb atom bonded to two Sb atoms. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

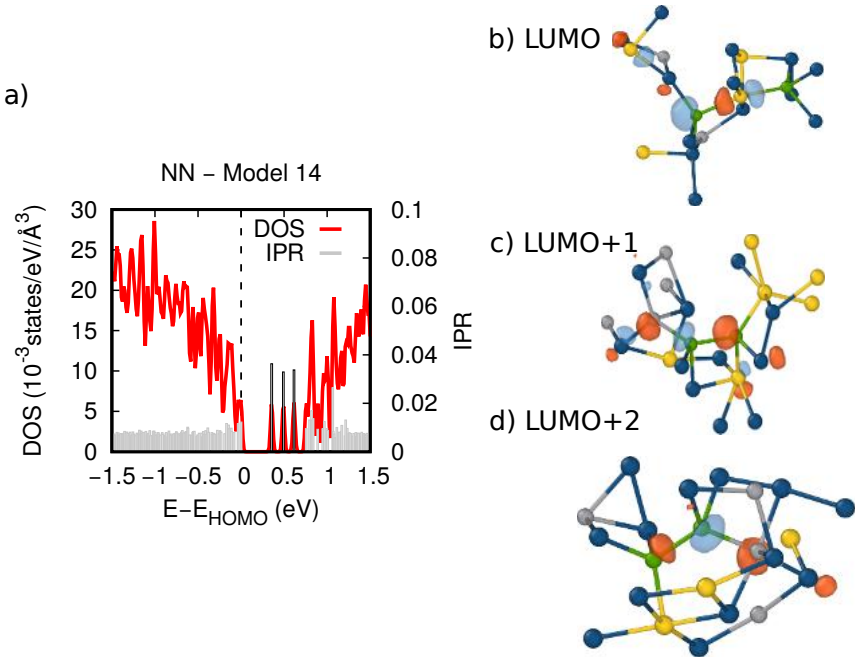

Figure S14: a) A zoom of the electronic DOS of model NN-14 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, c, and d. b) The LUMO is localized on a 5-coordinated Sb bonded to two Ge_{Td} atoms. The LUMO+1 (panel c) and the LUMO+2 (panel d) are localized on a chain of Ge atoms with two Ge_{Td} atoms. Ge, Ge_{Td} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

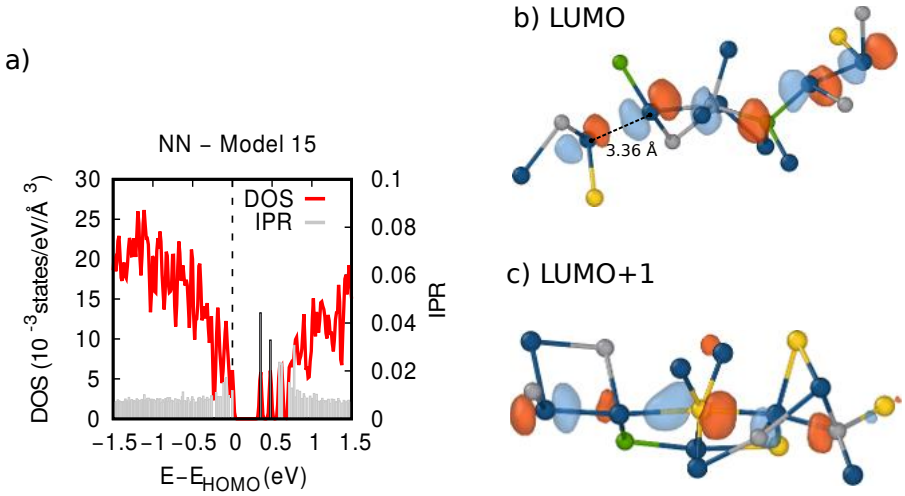

Figure S15: a) A zoom of the electronic DOS of model NN-15 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, and c. b) The LUMO is localized on long chain of wrong bonds. c) LUMO+1 is localized close to a 6-coordinated Sb atom, a Te-Te wrong bond and a 4-coordinated Te atom forming a Te-Te bond. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

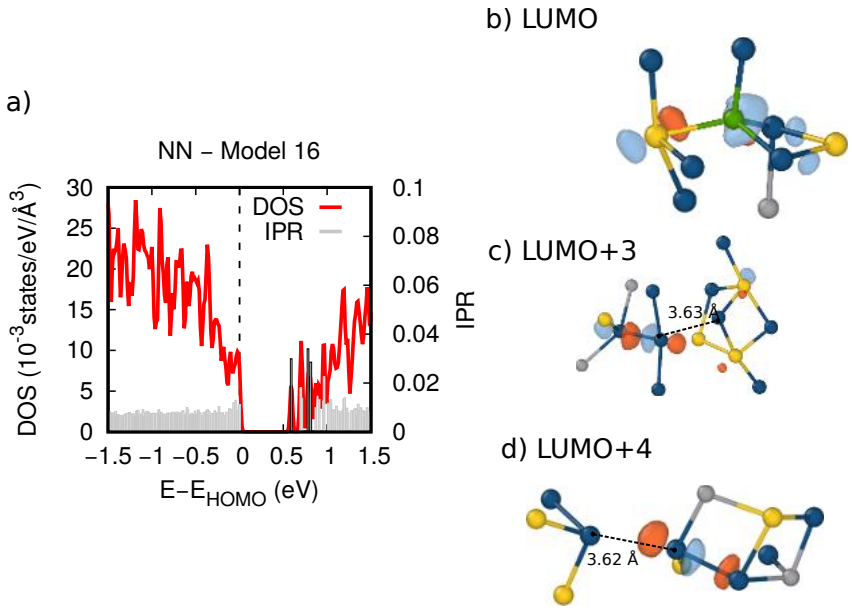

Figure S16: a) A zoom of the electronic DOS of model NN-16 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, c and d. b) The LUMO is localized on a Ge_T atom. LUMO+3 (panel c) and LUMO+4 (panel d) are localized on a 3-coordinated Te atom forming Te-Te wrong bond and a long Te-Te contact. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

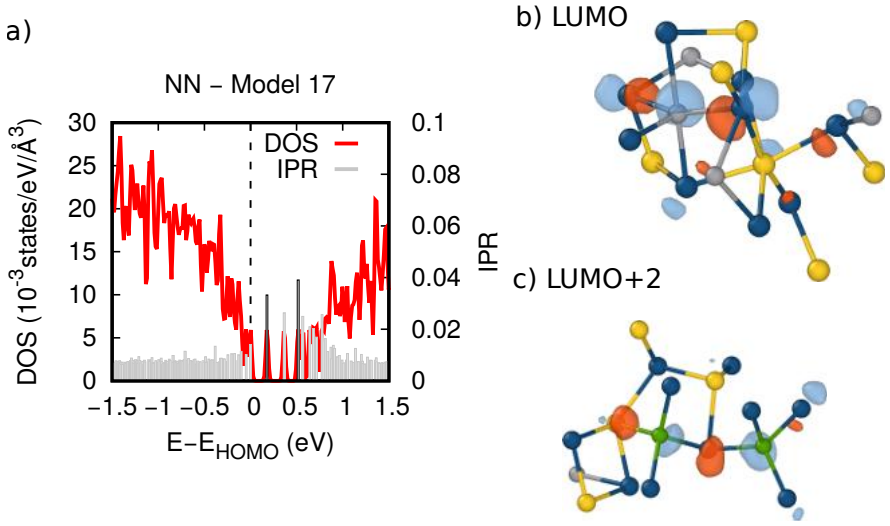

Figure S17: a) A zoom of the electronic DOS of model NN-17 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on 5-coordinated Ge atom. c) LUMO+2 is localized on a chain of two Ge_{tet} and a Ge-Sb wrong bond. Ge, Ge_{tet} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

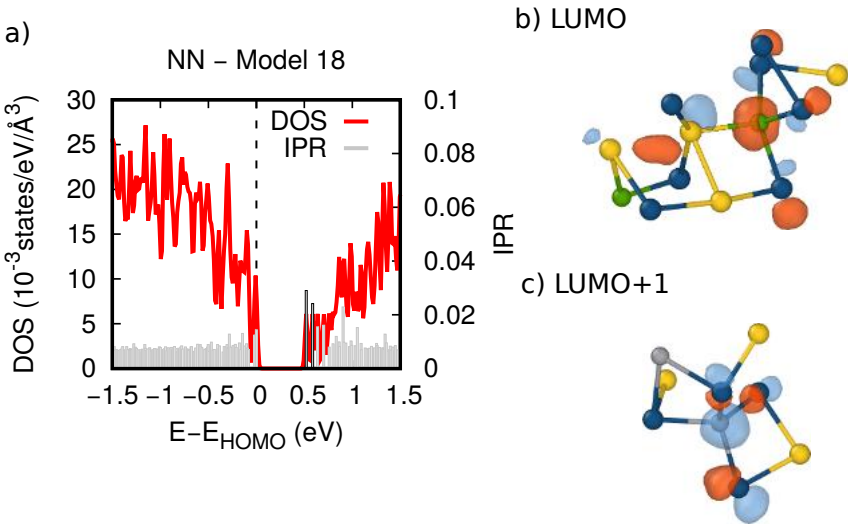

Figure S18: a) A zoom of the electronic DOS of model NN-18 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on a Ge_T atom with a wrong bond. c) LUMO+1 is localized on a 4-coordinated Ge atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

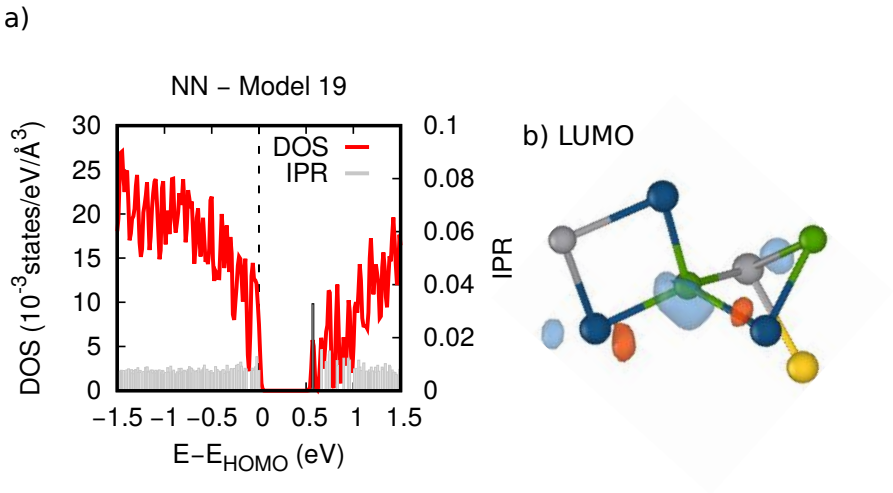

Figure S19: a) A zoom of the electronic DOS of model NN-19 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO is localized close to a Ge_{TV} atom with a wrong homopolar bond. Ge, Ge_{TV}, Sb and Te are depicted with gray, green, yellow and blue spheres.

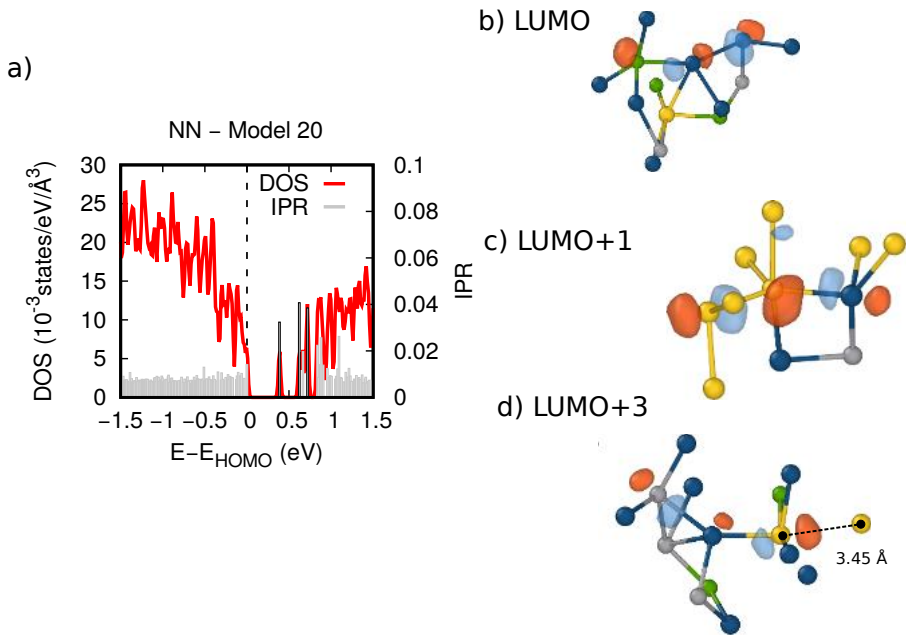

Figure S20: a) A zoom of the electronic DOS of model NN-20 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, c and d. b) The LUMO is localized on a Te atom forming a homopolar bond and a bond with a Ge_T atom. c) LUMO+1 is localized near a 5-coordinated Sb atom forming homopolar bonds. d) LUMO+3 is localized close to a 4-coordinated Sb forming a long contact with an Sb atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

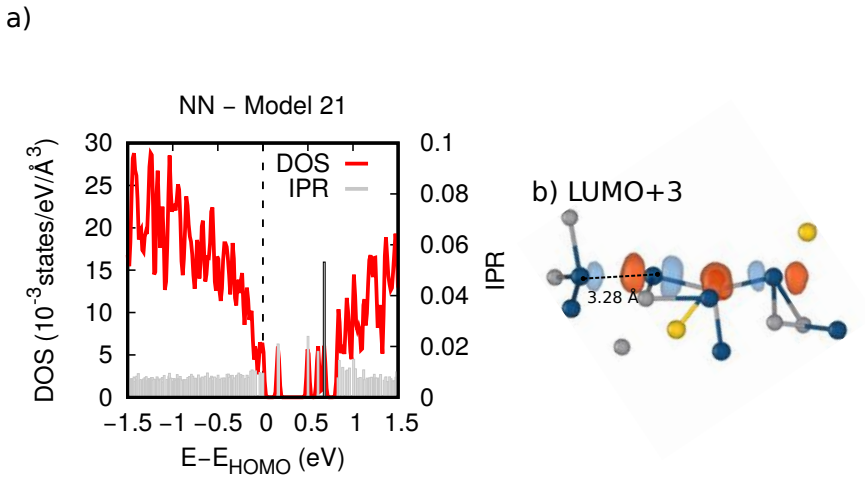

Figure S21: a) A zoom of the electronic DOS of model NN-21 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO+3 is localized close to a 5-coordinated Ge atom bonded to a Te atom forming in turn a long homopolar contact. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

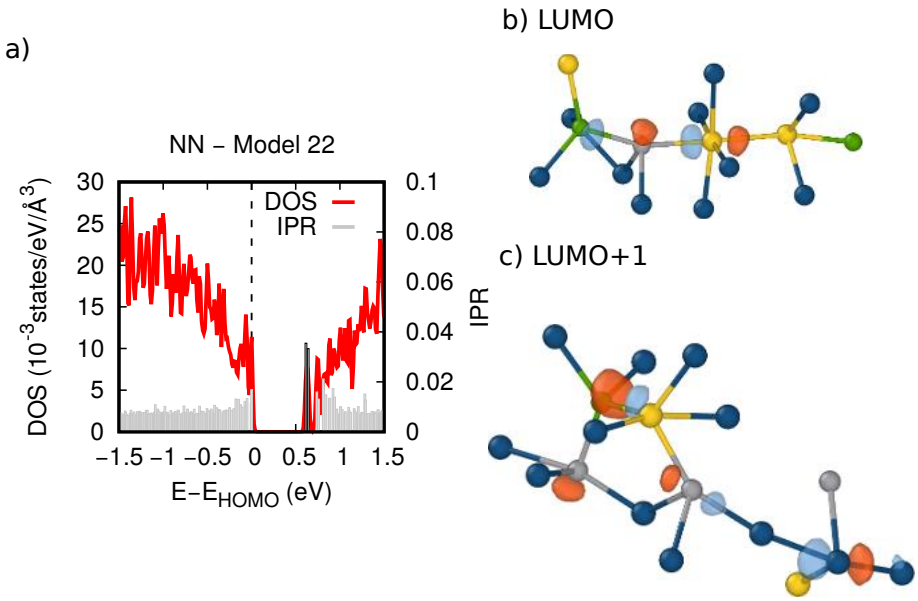

Figure S22: a) A zoom of the electronic DOS of model NN-22 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. LUMO (panel b) and LUMO+1 (panel c) are both localized on a long chain of wrong bonds. Ge$_{IV}$ (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

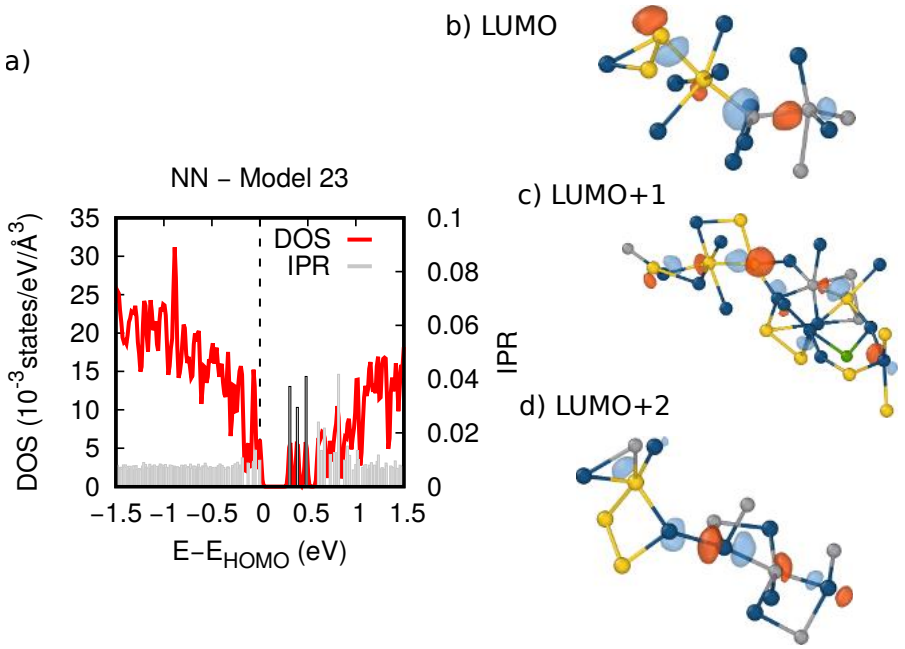

Figure S23: a) A zoom of the electronic DOS of model NN-23 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, c and d. LUMO (panel b) and LUMO+1 (panel c) are both localized on a long chain of wrong bonds. d) LUMO+2 is localized close to a 5-coordinated Ge atom bonded to a Te atom forming in turn a homopolar bond. Ge, Ge$_T$ (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

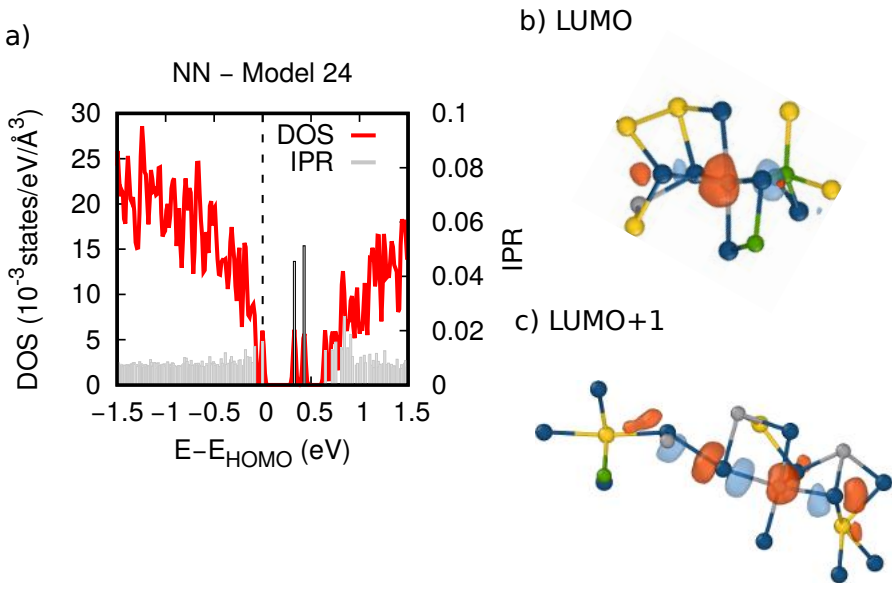

Figure S24: a) A zoom of the electronic DOS of model NN-24 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized close to a 6-coordinated Ge atom forming Ge-Ge_{Td} bond. c) LUMO+1 is localized close to a 5-coordinated Ge atom bonded to a Te atom forming in turn a homopolar bond. Ge, Ge_{Td} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

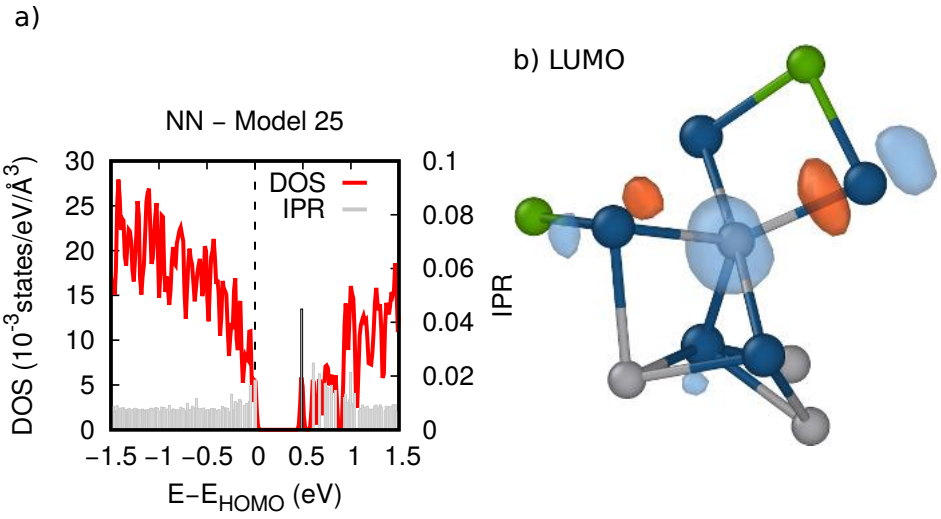

Figure S25: a) A zoom of the electronic DOS of model NN-26 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO is localized close to a 5-coordinated Ge atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

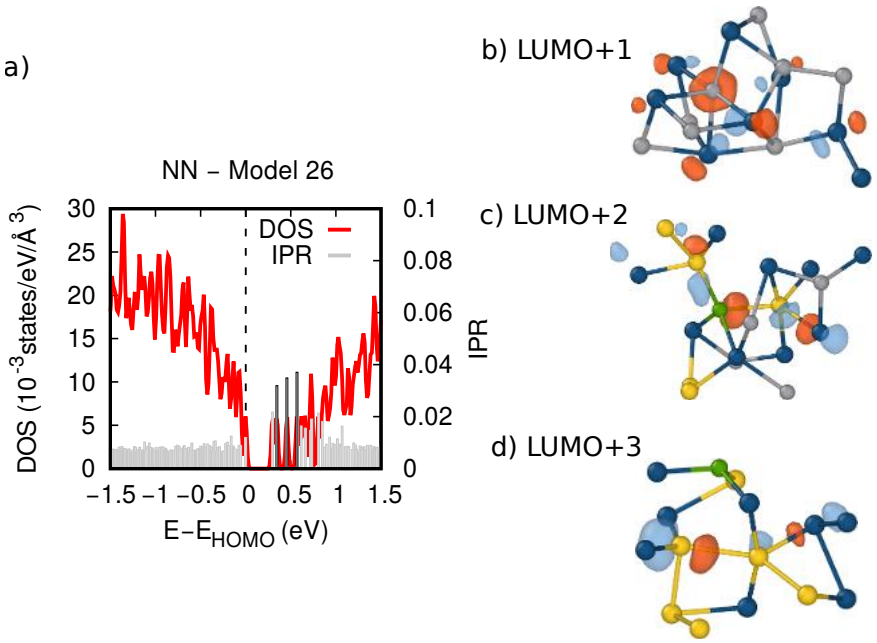

Figure S26: a) A zoom of the electronic DOS of model NN-26 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b, c and d. b) The LUMO+1 state is localized close to a 5-coordinated Ge atom. c) LUMO+2 is localized close to a Ge_T atom bonded to two Sb atoms. d) LUMO+3 is localized close to a 5-coordinated Sb atom forming homopolar bonds. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

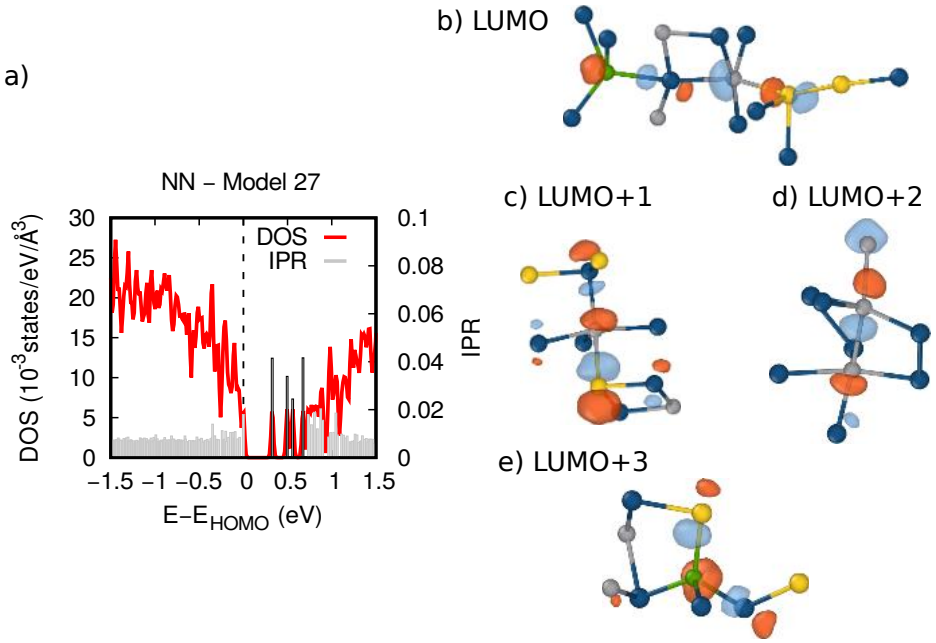

Figure S27: a) A zoom of the electronic DOS of model NN-27 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b)-e). b) LUMO is localized along a chain of wrong bonds involving Ge_{Td} atom and an overcoordinated Ge atom. c) LUMO+1 is localized on a a 5-coordinated Ge atom with a wrong bond. d) LUMO+2 is localized on a short chain of Ge atoms. e) LUMO+3 is localized close to two Ge_{Td} atoms forming a wrong bond with an Sb atom. Ge, Ge_{Td} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

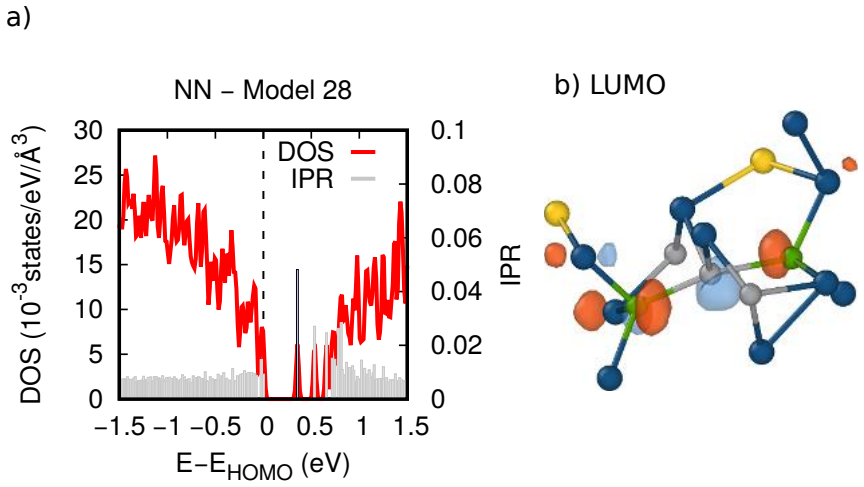

Figure S28: a) A zoom of the electronic DOS of model NN-28 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO state is localized on a chain of Ge atoms involving two Ge_T atoms. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

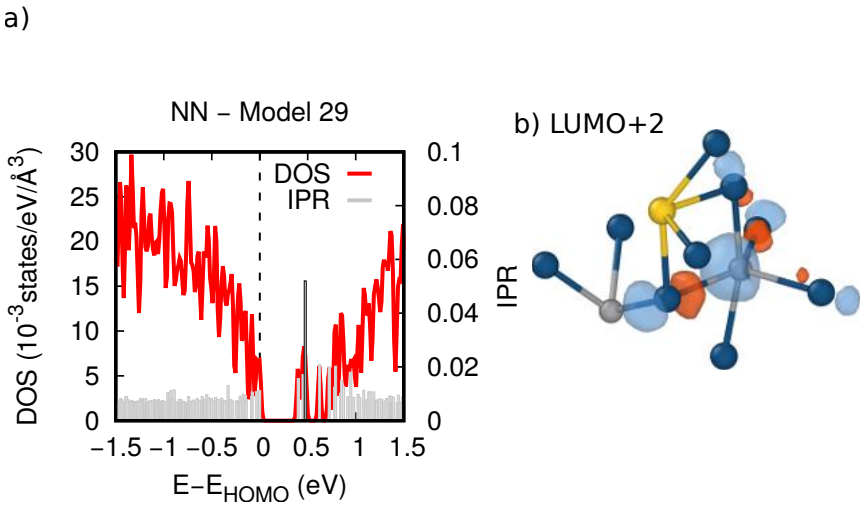

Figure S29: a) A zoom of the electronic DOS of model NN-29 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO state is localized close to a 5-coordinated Ge atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

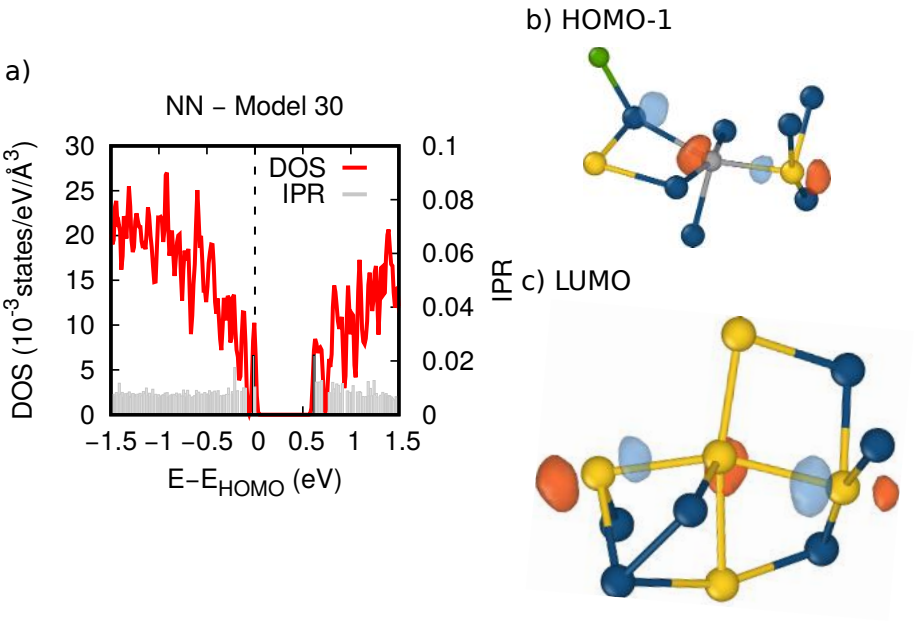

Figure S30: a) A zoom of the electronic DOS of model NN-30 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The HOMO-1 state is localized close to a 5-coordinated Ge atom forming a wrong Ge-Sb bond. c) LUMO+2 is localized on a chain of Sb atoms. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

2 Electronic DOS of DFT models

In Figs. S31-S40 we show the electronic density of states (DOS) computed with the HSE hydrid functional [1] of the ten 216-atom amorphous models of GST generated by quenching the melt in DFT-MD with the PBE functional [2] and then geometry-optimized at zero temperature with the HSE functional. Isosurfaces of the most localized in-gap states analyzed in the article are also shown (see Table S2).

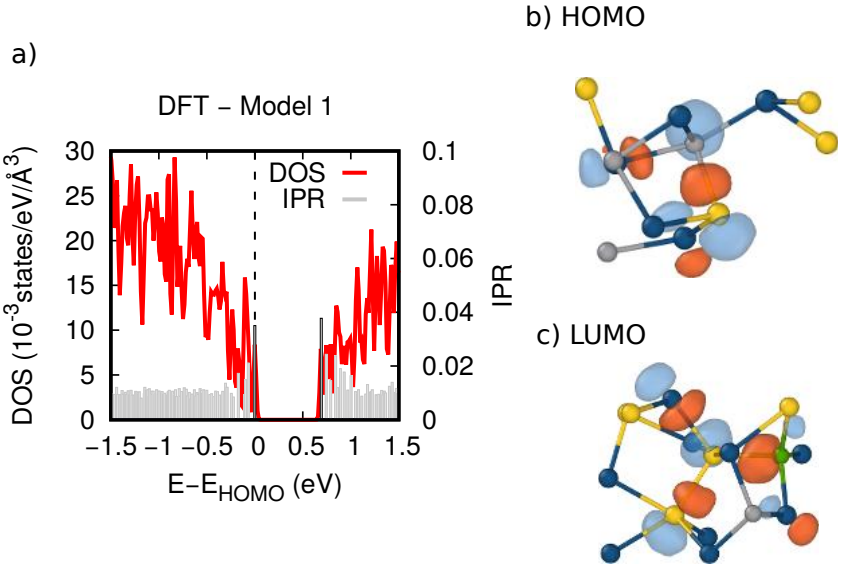

Figure S31: a) A zoom of the electronic DOS of model DFT-1 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The HOMO state is localized close to a 4-coordinated Ge atom forming a wrong bond with a 3-coordinated Sb atom. c) LUMO is localized on a 5-coordinated Sb atom forming wrong bonds with Sb and with Ge_T atoms. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

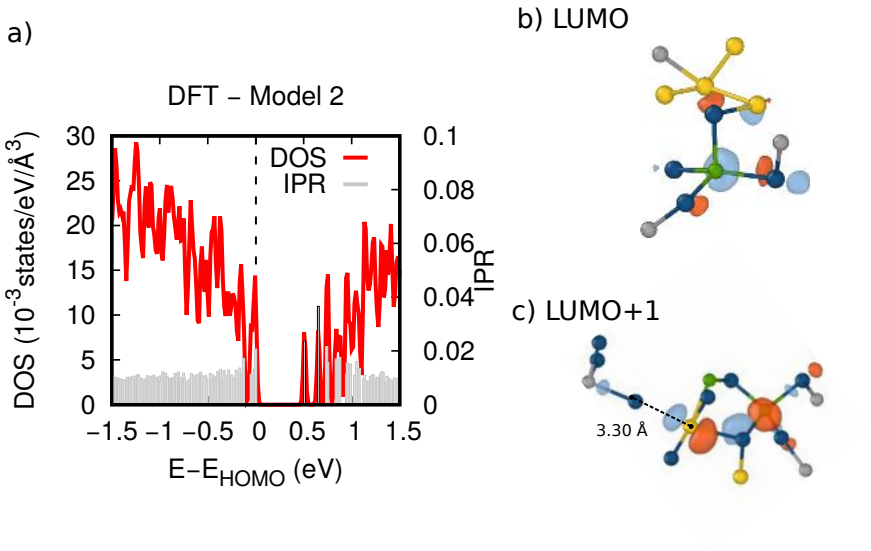

Figure S32: a) A zoom of the electronic DOS of model DFT-2 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on a Ge_{tet} atom. c) LUMO+1 is localized close to a Ge_{tet} atom and a 4-coordinated Sb atom forming wrong bond with Ge_{tet} atoms and a long contact with Te atom. Ge, Ge_{tet} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

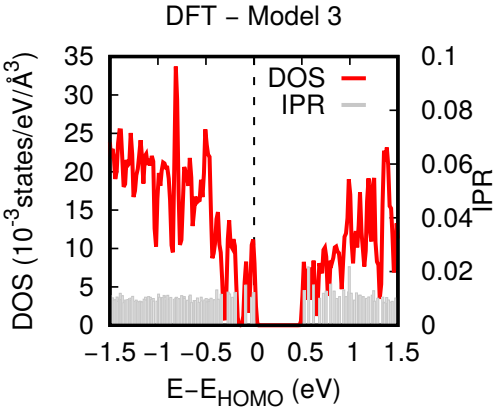

Figure S33: a) A zoom of the electronic DOS of model DFT-3 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). This model dose not display any localized states neither deep nor close to the band edges.

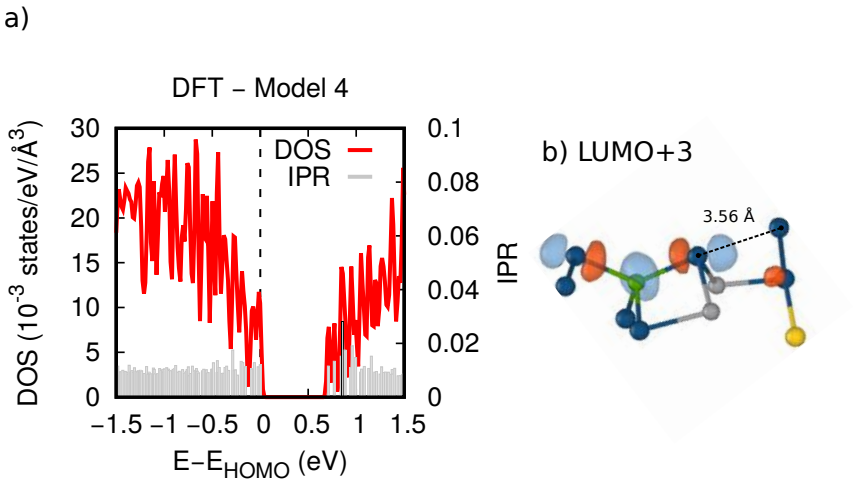

Figure S34: a) A zoom of the electronic DOS of model DFT-4 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO+3 state is localized close to a Ge_T atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

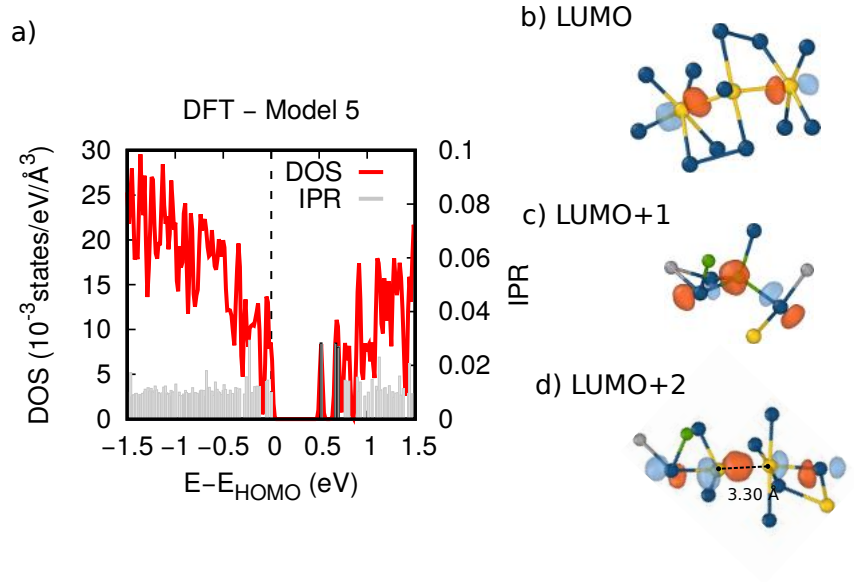

Figure S35: a) A zoom of the electronic DOS of model DFT-5 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on a short chain of Sb atoms. c) LUMO+1 is localized close to a Ge_T atom. d) LUMO+2 is localized close to a 5-coordinated Sb atom forming long contact with a Sb atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

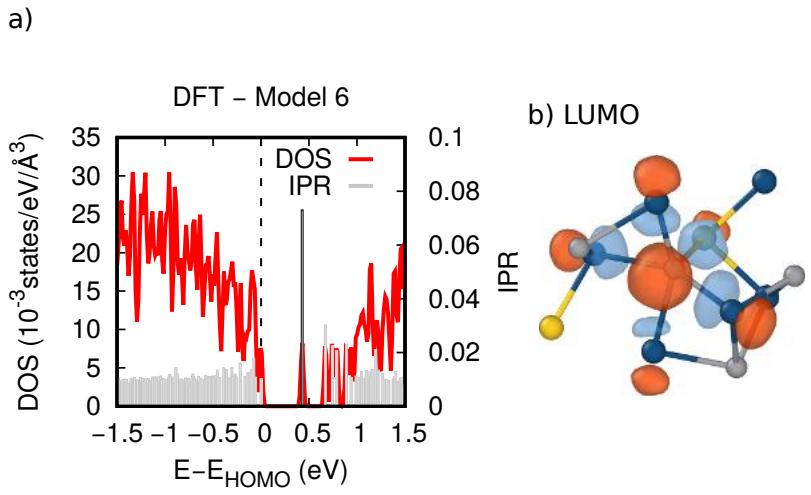

Figure S36: a) A zoom of the electronic DOS of model DFT-6 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of the state with highest IPR (highlighted with black border) is shown in panel b. b) The LUMO state is localized close to a 5-coordinated Ge atom. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

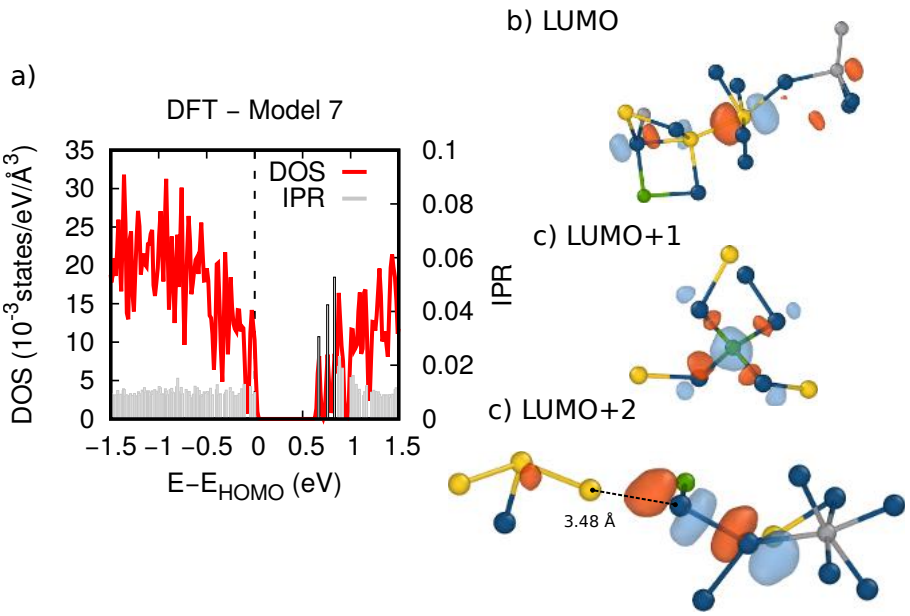

Figure S37: a) A zoom of the electronic DOS of model DFT-7 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on a homopolar Sb-Sb bond. c) LUMO+1 is localized an a Ge_T atom. d) LUMO+2 is localized on a Te-Te homopolar bond. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

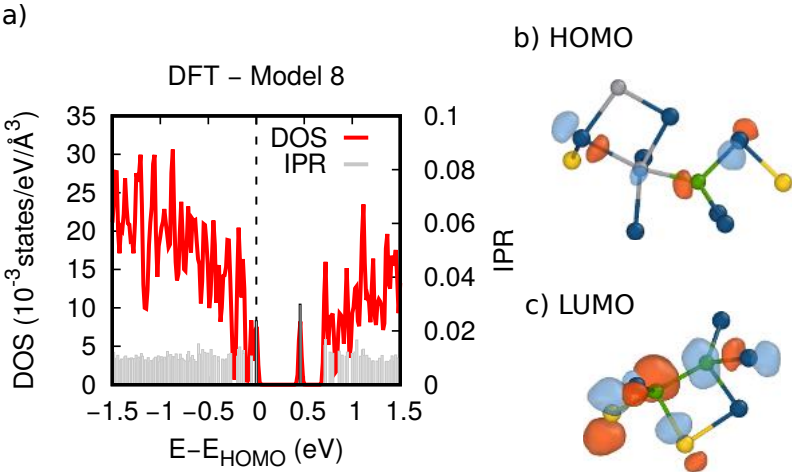

Figure S38: a) A zoom of the electronic DOS of model DFT-8 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The HOMO is localized on a homopolar Ge-Ge$_{tet}$ bond. c) LUMO is localized close to two bonded Ge$_{tet}$ atoms. Ge, Ge$_{tet}$ (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

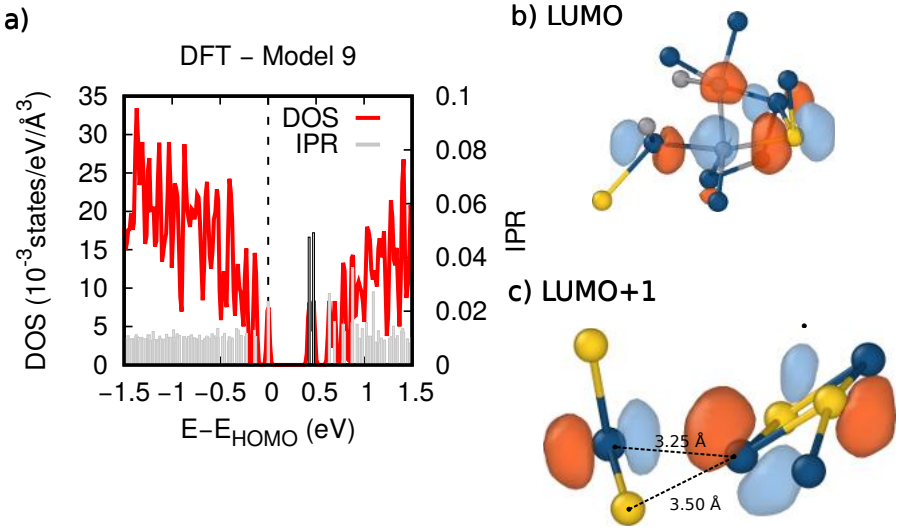

Figure S39: a) A zoom of the electronic DOS of model DFT-9 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b and c. b) The LUMO is localized on a 5-coordinated Ge atom forming a bond with undercoordinated Sb atom and another 5-coordinated Ge atom. c) LUMO+1 is localized on two Te atoms with long Te-Te and Te-Sb contacts. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

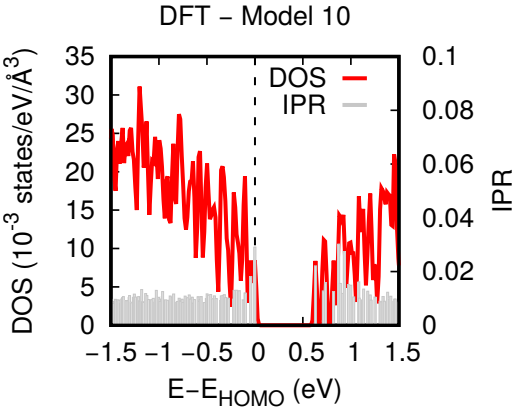

Figure S40: a) A zoom of the electronic DOS of model DFT-10 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero of energy is the highest occupied state (HOMO). This model dose not display any localized states neither deep nor close to the band edges.

3 Electronic DOS of 459-atom models

In Figs. S41-S44 we show the electronic density of states (DOS) computed with the HSE hydrid functional [1] of the four 459-atom amorphous models of GST generated by quenching the melt in DFT-MD in Ref. [3] and then geometry-optimized at zero temperature with the HSE functional. Isosurfaces of the most localized in-gap states analyzed in the article are also shown (see Table S2).

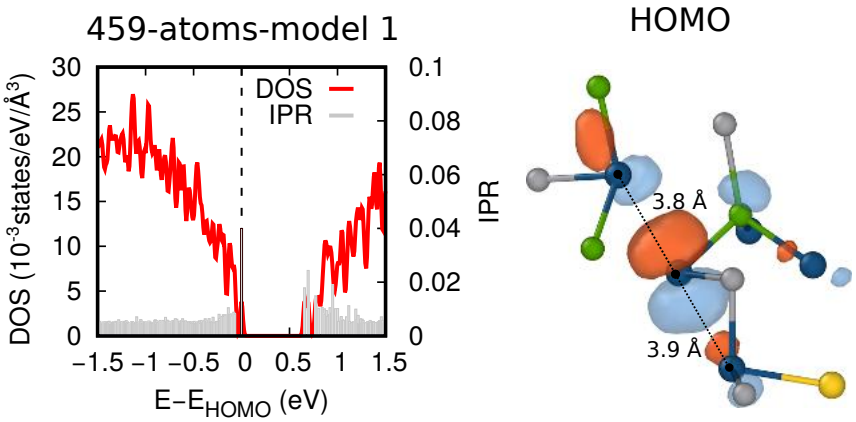

Figure S41: a) A zoom of the electronic DOS of 459-atom model 1 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b. b) The HOMO is localized on a 2-coordinated Te atom forming two long Te-Te contacts. Ge, Ge$_T$ (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

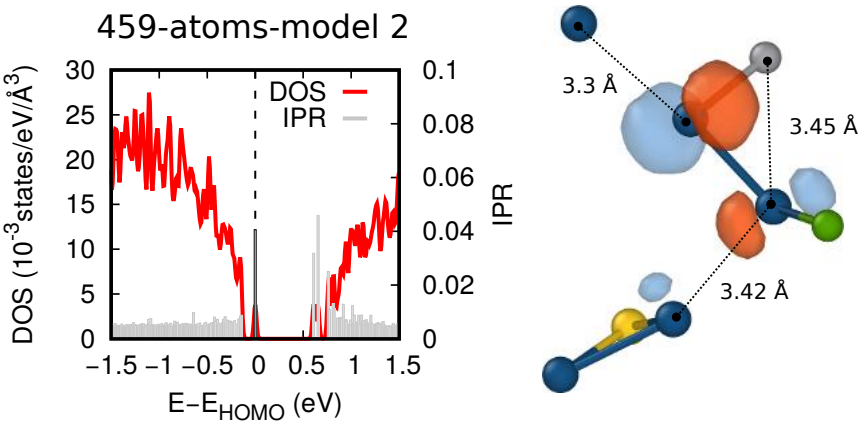

Figure S42: a) A zoom of the electronic DOS of 459-atom model 2 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b. b) The HOMO is localized on a 2-coordinated Te forming two long Te-Te contacts. Ge, Ge_T (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

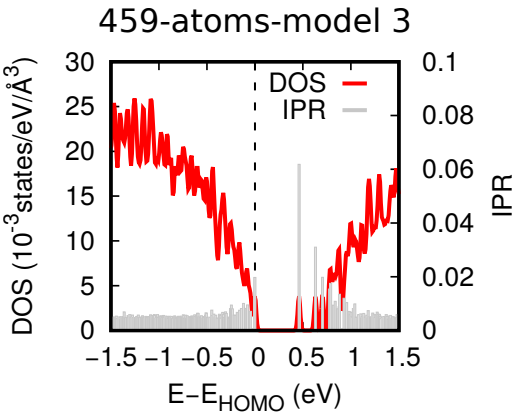

Figure S43: a) A zoom of the electronic DOS of 459-atom model 3 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero energy is the highest occupied state (HOMO).

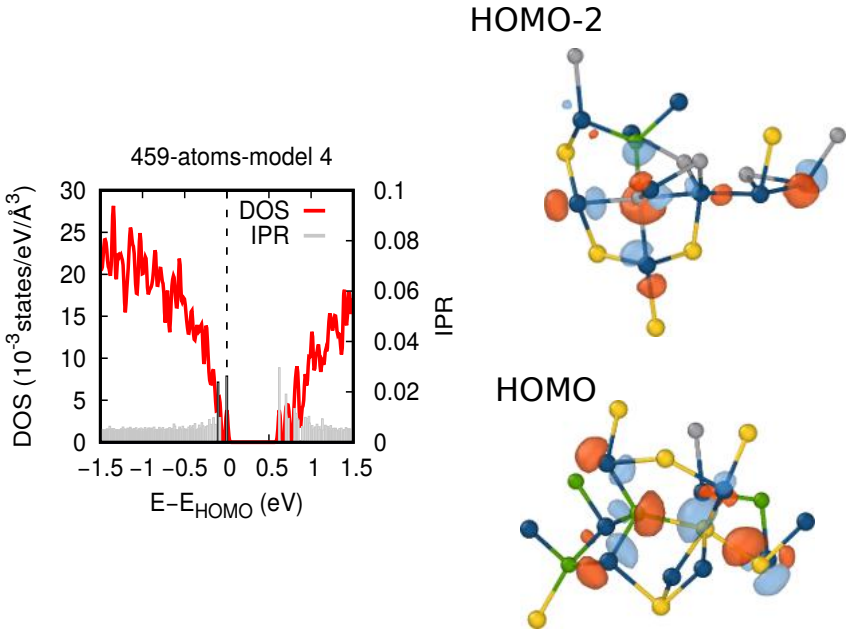

Figure S44: a) A zoom of the electronic DOS of 459-atom model 4 (red curve) close to the band gap and the value of the IPR (gray spikes) of KS states at the Γ point. The zero energy is the highest occupied state (HOMO). The isosurfaces of states with high IPR (highlighted with black border) are shown in panels b-c. b) HOMO-2 state is localized on an overcoordinated Ge atom bonded to a short chain of Te atoms. c) The HOMO is localized on a short chain of wrong bonds involving Sb-Sb and Sb-Ge_{γ} bonds. Ge, Ge_{γ} (tetrahdral Ge), Sb and Te are depicted with gray, green, yellow and blue spheres.

4 Validation of the electronic properties of NN models

In this section, we report the electronic DOS (see Figs. S45-47) of models generated to benchmark electronic properties of NN models against DFT models.

In Fig. S45 the electronic DOS of four 216-atom NN models (sNN-1, sNN-2, sNN-3 and sNN-4) are compared with the DOS averaged over the DFT models with the same size. The DOS of these smaller NN models have no in-gap states in the energy range 0.1-0.3 eV (HOMO at zero energy) as opposed to the larger 297-atom NN models.

We then verified that the states in the energy range 0.1-0.3 eV observed in the larger NN models are not an artifact of the NN potential, by performing a hybrid quench (NN from 1000 K to 600 K, then DFT down to 300 K) or a hybrid annealing

(full quench followed by first annealing with NN-MD, then a second annealing with DFT-MD at 300 K). In Fig. S46 and S47 we report the electronic DOS of models NN-3 and NN-7 subjected to this protocol that still feature in-gap states in the 0.1-0.3 eV range.

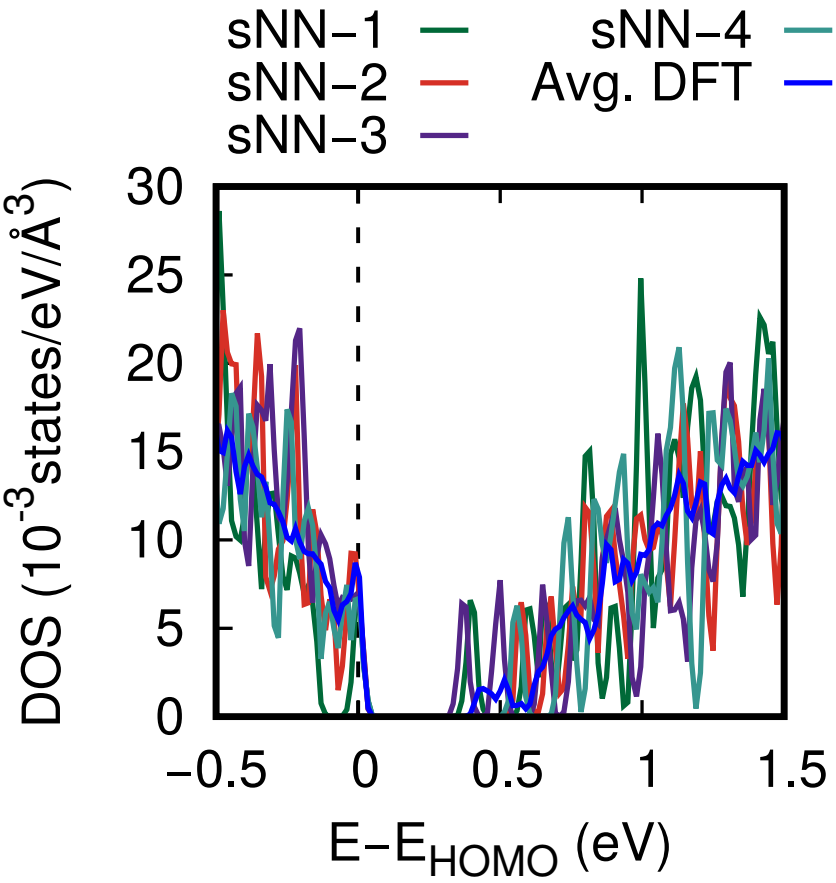

Figure S45: a) A zoom close to the band gap of the electronic DOS of sNN models (red curve) compared with the DOS averaged over the DFT models (blue curve).

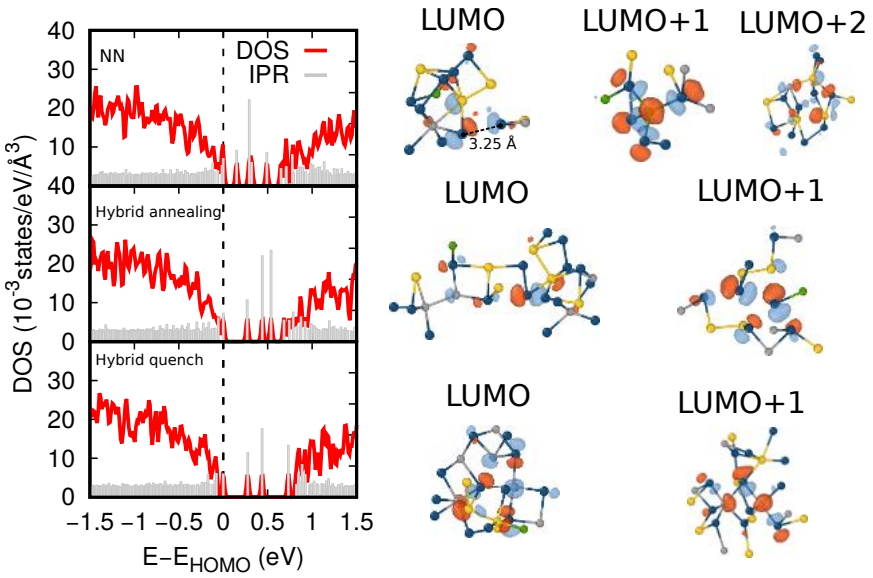

Figure S46: a) A zoom close to the band gap of the electronic DOS of model NN-3: Full NN quench (upper panel), hybrid NN+DFT-MD annealing at 300 K (central panel) and hybrid quench NN+DFT-NN (lower panel, see article). Isosurfaces of in-gap states below 0.35 eV are shown on the left.

5 Mobility gap and energy position of in-gap states

In this section we report the mobility gap of all 40 models (Table S1) and the position in energy of all in-gap states analyzed in the article (Table S2).

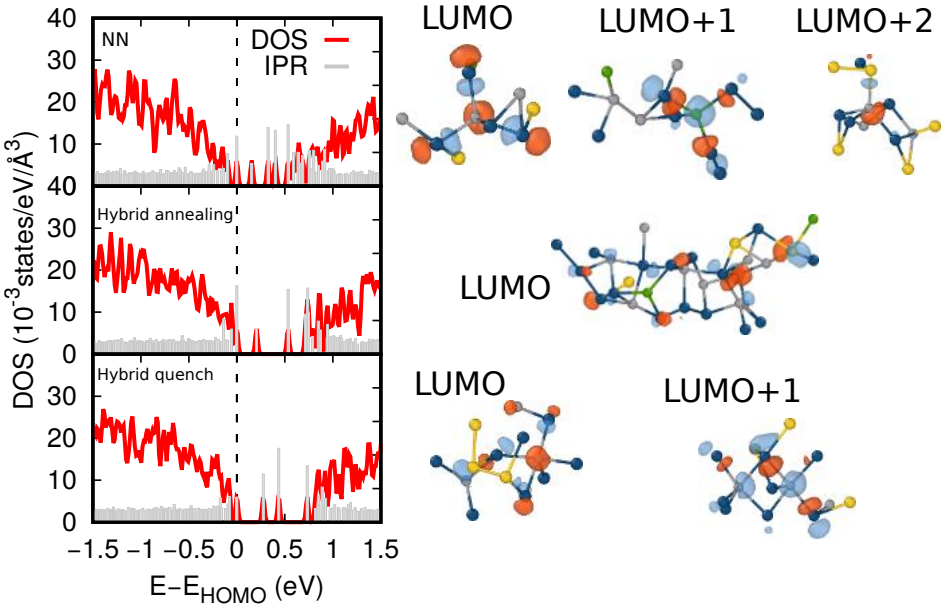

Figure S47: a) A zoom close to the band gap of the electronic DOS of model NN-7: Full NN quench (upper panel), hybrid NN+DFT-MD annealing at 300 K (central panel) and hybrid quench NN+DFT-NN (lower panel, see article). Isosurfaces of in-gap states below 0.35 eV are shown on the left.

Table S1: Mobility gap (eV) of the different models (generated by NN or DFT molecular dynamics) assigned by identifying the VB edge as the energy below which all states have a IPR lower than 0.018 and are therefore delocalized and similarly for the CB edges. Due the small size of our simulation cells this procedure is subject to some arbitrariness and uncertainties.

NN Model	Mobility gap (eV)	DFT Model	Mobility gap (eV)
NN-1	0.74	DFT-1	0.85
NN-2	0.892	DFT-2	0.74
NN-3	0.659	DFT-3	0.54
NN-4	0.81	DFT-4	0.71
NN-5	0.8	DFT-5	0.77
NN-6	0.78	DFT-6	0.74
NN-7	0.78	DFT-7	0.86
NN-8	0.69	DFT-8	0.76
NN-9	0.89	DFT-9	0.81
NN-10	0.96	DFT-10	0.67
NN-11	0.65		
NN-12	0.75		
NN-13	0.79		
NN-14	0.75		
NN-15	0.71		
NN-16	0.85		
NN-17	0.71		
NN-18	0.64		
NN-19	0.67		
NN-20	0.85		
NN-21	0.83		
NN-22	0.74		
NN-23	0.64		
NN-24	0.64		
NN-25	0.78		
NN-26	0.61		
NN-27	0.73		
NN-28	0.71		
NN-29	0.78		
NN-30	0.74		

Table S2: Position in energy (eV) with respect to the HOMO of the 84 in-gap states in the different models (see text) whose localization properties is analyzed in the article.

VB state	Energy (eV)	Deep state	Energy (eV)	CB state	Energy (eV)
NN-1 HOMO-2	-0.1127	NN-1 LUMO	0.2743	NN-1 LUMO+1	0.5383
NN-6 LUMO	0.136	NN-2 LUMO+4	0.3174	NN-1 LUMO+2	0.6098
NN-7 HOMO	0	NN-2 LUMO+5	0.6533	NN-6 LUMO+1	0.6166
NN-10 HOMO	0	NN-3 LUMO+1	0.2925	NN-7 LUMO+3	0.5333
NN-17 LUMO	0.179	NN-4 LUMO	0.2401	NN-8 LUMO+3	0.4997
NN-30 HOMO-1	-0.0156	NN 4-LUMO+1	0.5545	NN-8 LUMO+5	0.6496
DFT-1 HOMO	0	NN-5 LUMO	0.2957	NN-11 LUMO	0.3457
DFT-8 HOMO	0	NN-5 LUMO+2	0.4805	NN-14 LUMO+2	0.6149
459-1 HOMO	0	NN-7 LUMO+1	0.3248	NN-16 LUMO+3	0.7817
459-2 HOMO	0	NN-7 LUMO+2	0.4009	NN-16 LUMO+4	0.8126
459-3 HOMO	0	NN-8 LUMO+2	0.4761	NN-17 LUMO+2	0.517
459-4 HOMO	0	NN-9 LUMO	0.455	NN-18 LUMO	0.5197
459-4 HOMO-2	-0.103	NN-9 LUMO+1	0.6188	NN-18 LUMO+1	0.5861
		NN-10 LUMO	0.5331	NN-19 LUMO	0.572
		NN-12 LUMO	0.3598	NN-20 LUMO+3	0.7166
		NN-12 LUMO+1	0.3679	NN-21 LUMO+3	0.6809
		NN-13 LUMO	0.4392	NN-22 LUMO	0.6154
		NN-14 LUMO	0.356	NN-22 LUMO+1	0.6409
		NN-14 LUMO+1	0.4916	NN-23 LUMO+2	0.4812
		NN-15 LUMO	0.354	NN-26 LUMO+2	0.457
		NN-15 LUMO+1	0.4787	NN-26 LUMO+3	0.5684
		NN-16 LUMO	0.5903	NN-27 LUMO+2	0.5576
		NN-20 LUMO	0.387	NN-27 LUMO+3	0.6742
		NN-20 LUMO+1	0.6227	NN-30 LUMO	0.6164
		NN-23 LUMO	0.3128	DFT-1 LUMO	0.6917
		NN-23 LUMO+1	0.3905	DFT-2 LUMO+1	0.6527
		NN-24 LUMO	0.3279	DFT-4 LUMO+3	0.857
		NN-24 LUMO+1	0.428	DFT-5 LUMO+1	0.6728
		NN-25 LUMO	0.487	DFT-5 LUMO+2	0.6975
		NN-26 LUMO+1	0.3461	DFT-7 LUMO	0.6683
		NN-27 LUMO	0.3267	DFT-7 LUMO+1	0.7602
		NN-27 LUMO+1	0.4952	DFT-7 LUMO+2	0.8316
		NN-28 LUMO	0.355		
		NN-29 LUMO+2	0.474		
		DFT-2 LUMO	0.5081		
		DFT-5 LUMO	0.5194		
		DFT-6 LUMO	0.4288		
		DFT-8 LUMO	0.4562		
		DFT-9 LUMO	0.427		
		DFT-9 LUMO+1	0.4732		

References

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