Electronic Supporting information

The Gold-rich $R_3Au_7Sn_3$: Establishing the Interdependence between Electronic Features and Physical Properties

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Table S1. Distances, multiplicities and –ICOHP/bond for all homo- and heteroatomic interactions within the hexagonal $Gd_3Au_7Sn_3$.

Interaction	Distance [Å]	No. of bonds	−ICOHP/bond [eV/bond]	Interaction	Distance [Å]	No. of bonds	−ICOHP/bond [eV/bond]
Au-Au interactions				*Au-Gd interactions*			
Au1-Au2	2.960	12	0.9712	Au1-Gd3	2.956	6	0.8706
Au2-Au2	2.906	6	1.2693	Au2-Gd3	3.025	12	0.7788
Au2-Au2	3.147	6	0.6790	Au2-Gd3	3.082	12	0.7736
Au2-Au2	3.339	12	0.3824	Au2-Gd3	3.202	12	0.5638
Au2-Au2	4.467	12	−0.0219	Au2-Gd3	3.266	12	0.5423
Au-Sn interactions				*Sn-Gd interactions*			
Au1-Sn4	2.753	4	1.5987	Sn4-Gd3	3.338	12	0.4127
Au2-Sn4	2.845	12	1.3823	Sn4-Gd3	4.039	12	0.0742
Au2-Sn4	2.881	12	1.2218	Sn5-Gd3	3.463	12	0.4282
Au2-Sn5	2.952	12	1.2291				

Table S2. Distances, multiplicities and –ICOHP/bond for all homo- and heteroatomic interactions within the hexagonal $Y_3Au_7Sn_3$.

Interaction	Distance [Å]	No. of bonds	−ICOHP/bond [eV/bond]	Interaction	Distance [Å]	No. of bonds	−ICOHP/bond [eV/bond]
Au-Au interactions				*Au-Y interactions*			
Au1-Au2	2.962	12	0.9649	Au1-Y3	2.922	6	0.8530
Au2-Au2	2.903	6	1.2847	Au2-Y3	3.016	12	0.7416
Au2-Au2	3.188	6	0.6141	Au2-Y3	3.088	12	0.7242
Au2-Au2	3.296	12	0.4267	Au2-Y3	3.163	12	0.5589
Au2-Au2	4.472	12	−0.0210	Au2-Y3	3.250	12	0.5159
Au-Sn interactions				*Sn-Y interactions*			
Au1-Sn4	2.754	4	1.6051	Sn4-Y3	3.280	12	0.4256
Au2-Sn4	2.849	12	1.3724	Sn4-Y3	4.016	12	0.0759
Au2-Sn4	2.891	12	1.2024	Sn5-Y3	3.501	12	0.3678
Au2-Sn5	2.907	12	1.3230				

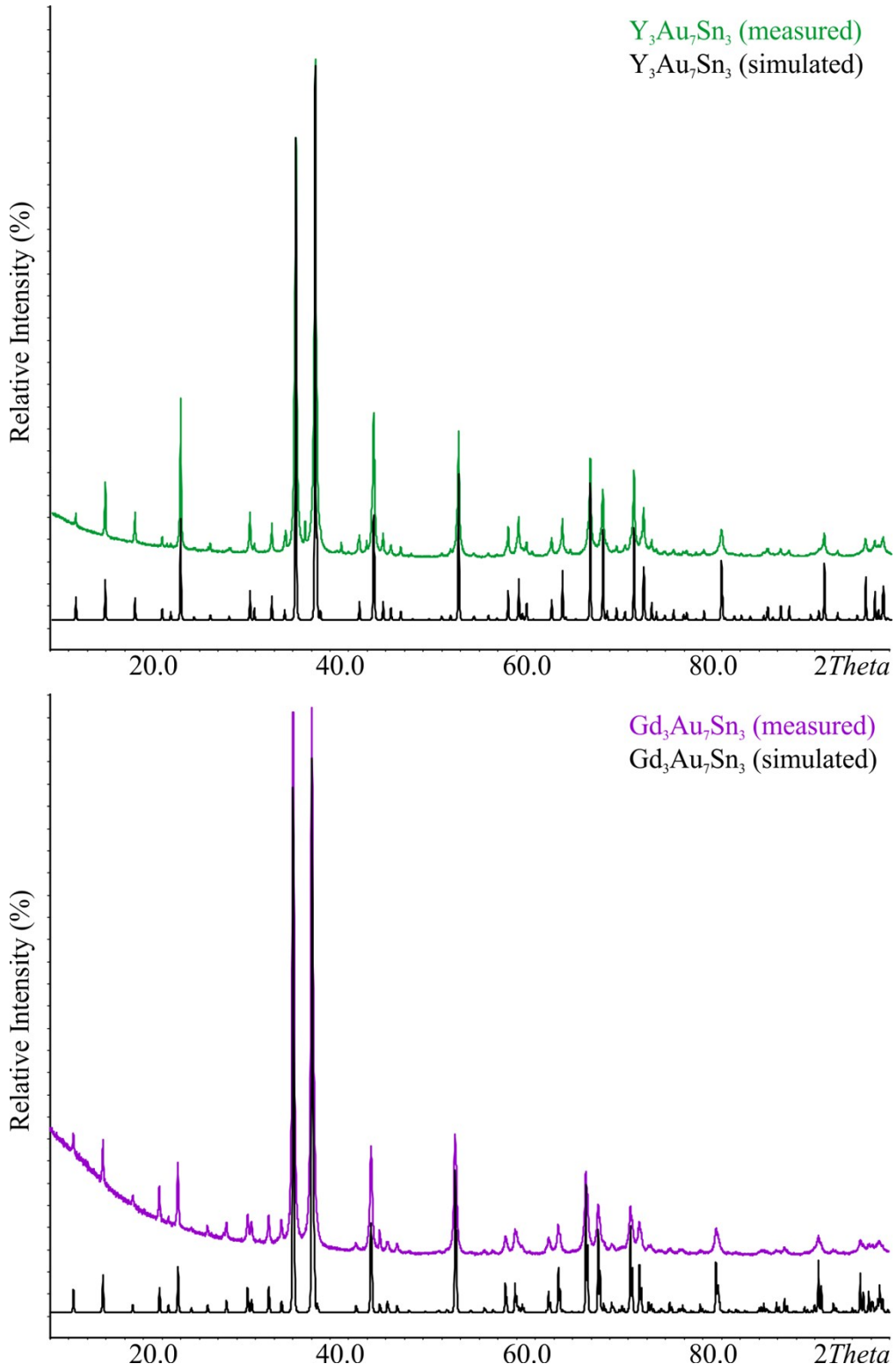

Figure S1. Measured and simulated powder X-ray diffraction patterns of $Y_3Au_7Sn_3$ and $Gd_3Au_7Sn_3$.

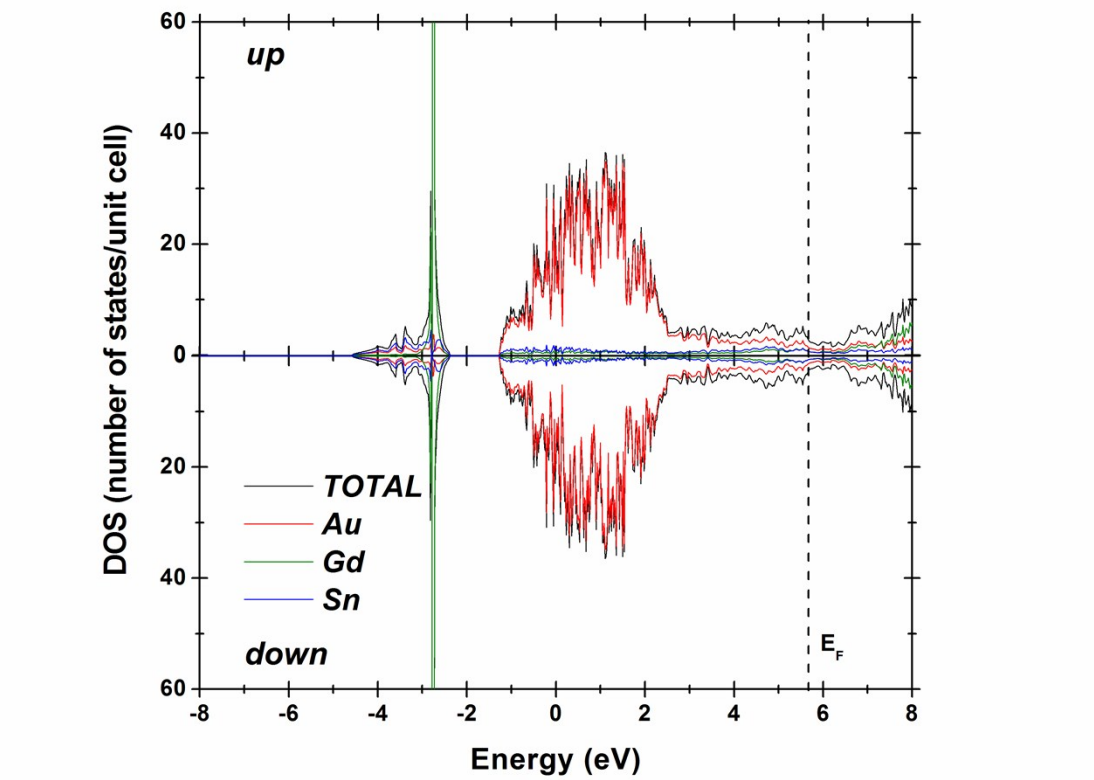

Figure S2. Plots of the spin-polarized total and projected DOS curves for the AFM spin ordering model 1. The Fermi level is represented by the dotted lines.

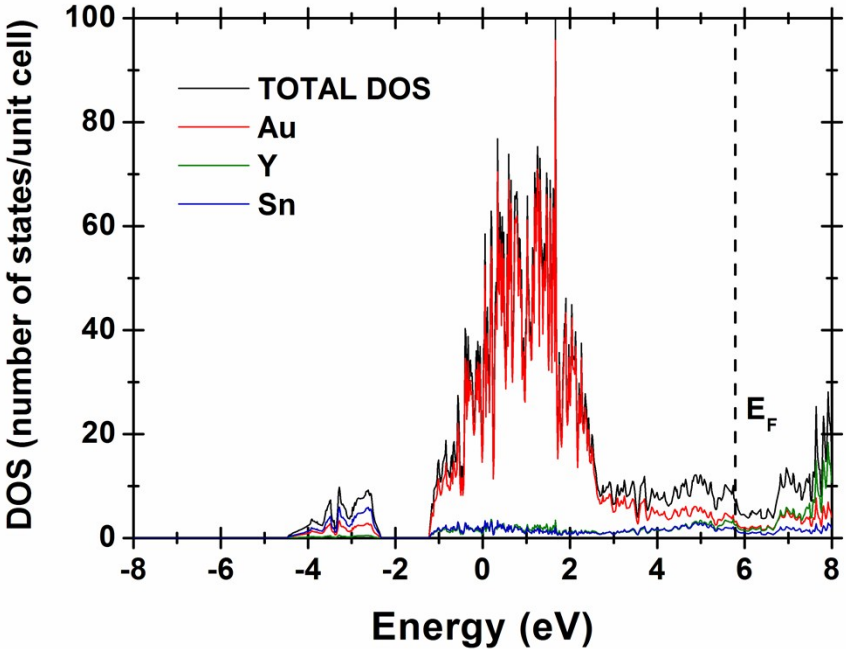

Figure S3. Plots of the total and projected DOS curves for $Y_3Au_7Sn_3$. The Fermi level is represented by the dotted lines.

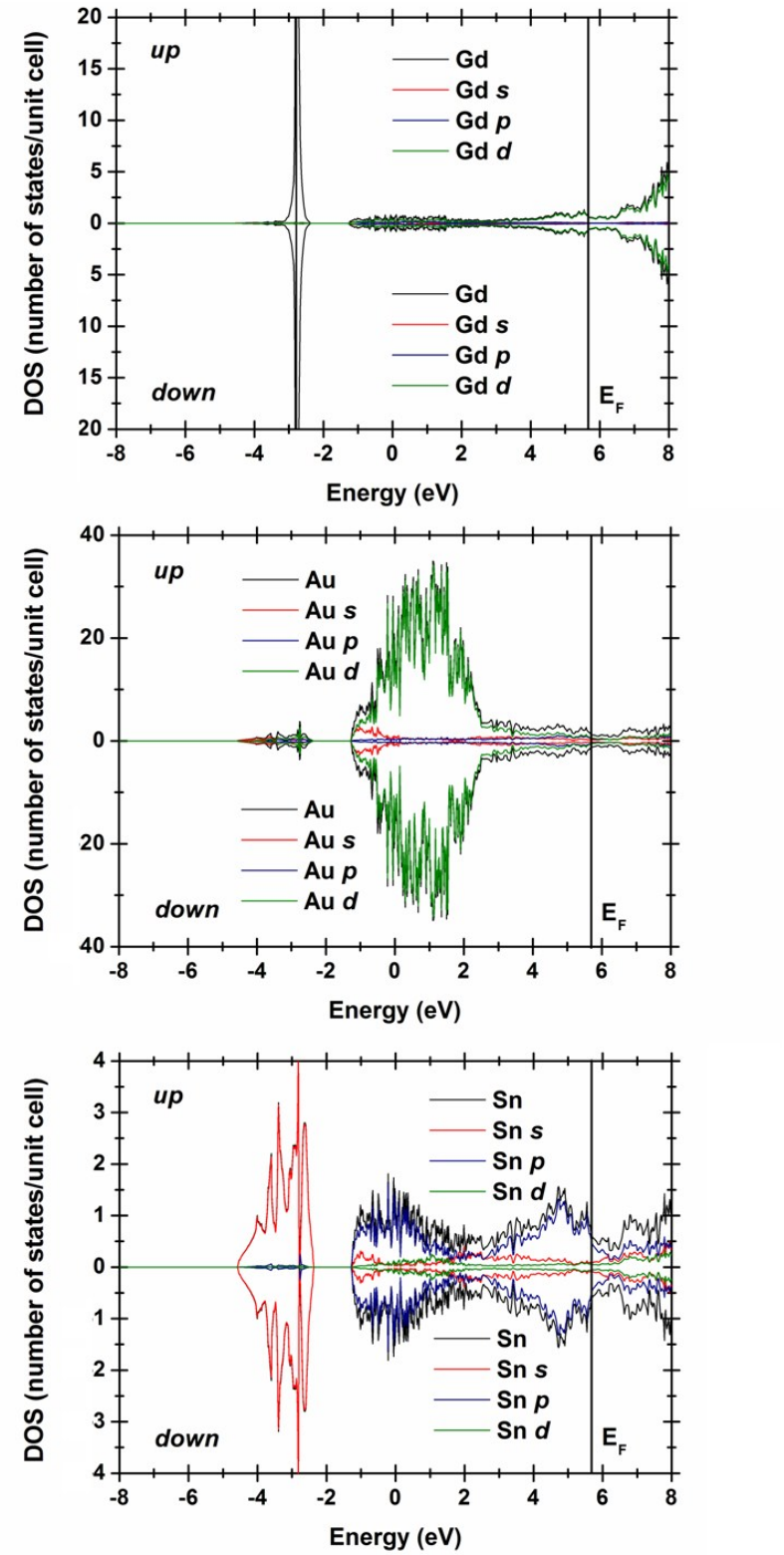

Figure S4. Site and orbital projected DOS (VASP) for the spin ordering model 1. The Fermi level is represented by the vertical lines.

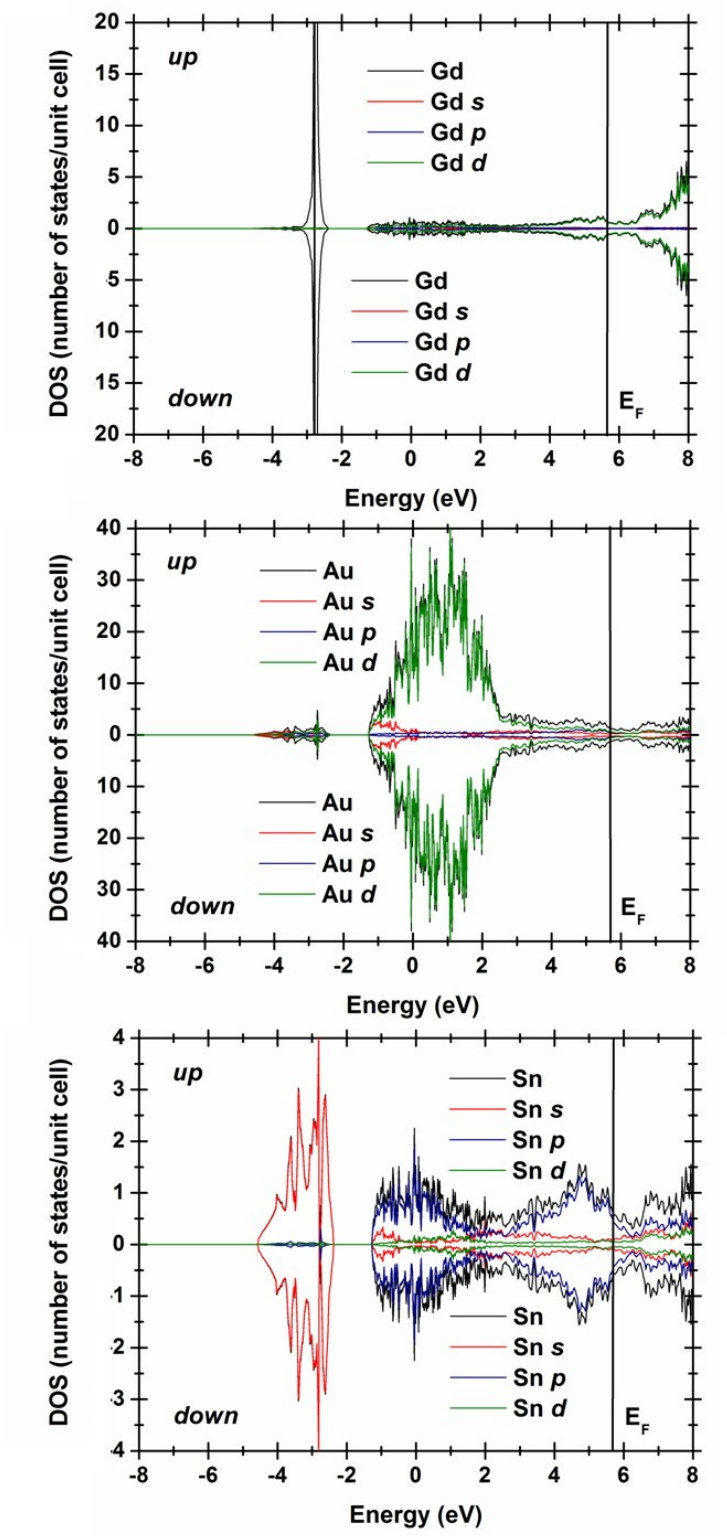

Figure S5. Site and orbital projected DOS (VASP) for the model 2. The Fermi level is represented by the vertical lines.

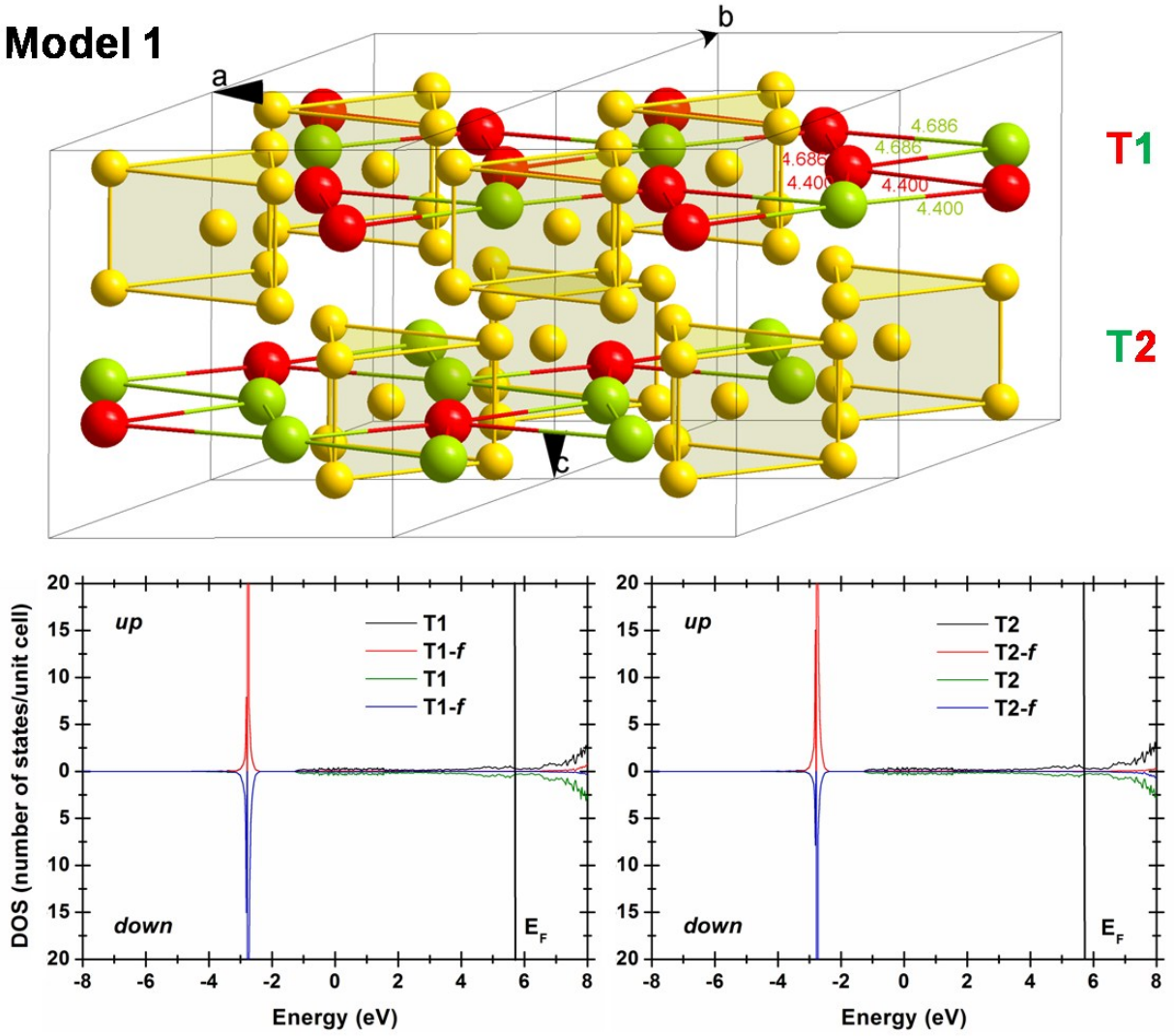

Figure S6. **Top**: Illustration of the antiferromagnetic spin ordering model 1 for the layers of the isosceles Gd triangles (T1 and T2). Spin-up gadolinium atoms are represented in green, while spin-down Gd atoms are highlighted in red. The Au@Au_6 clusters (yellow) are embedded within the Gd nets and Sn atoms are omitted for clarity. **Bottom**: T1 as well as T2 layers contribute to the spin-up and spin-down Gd *f* PDOS curves.

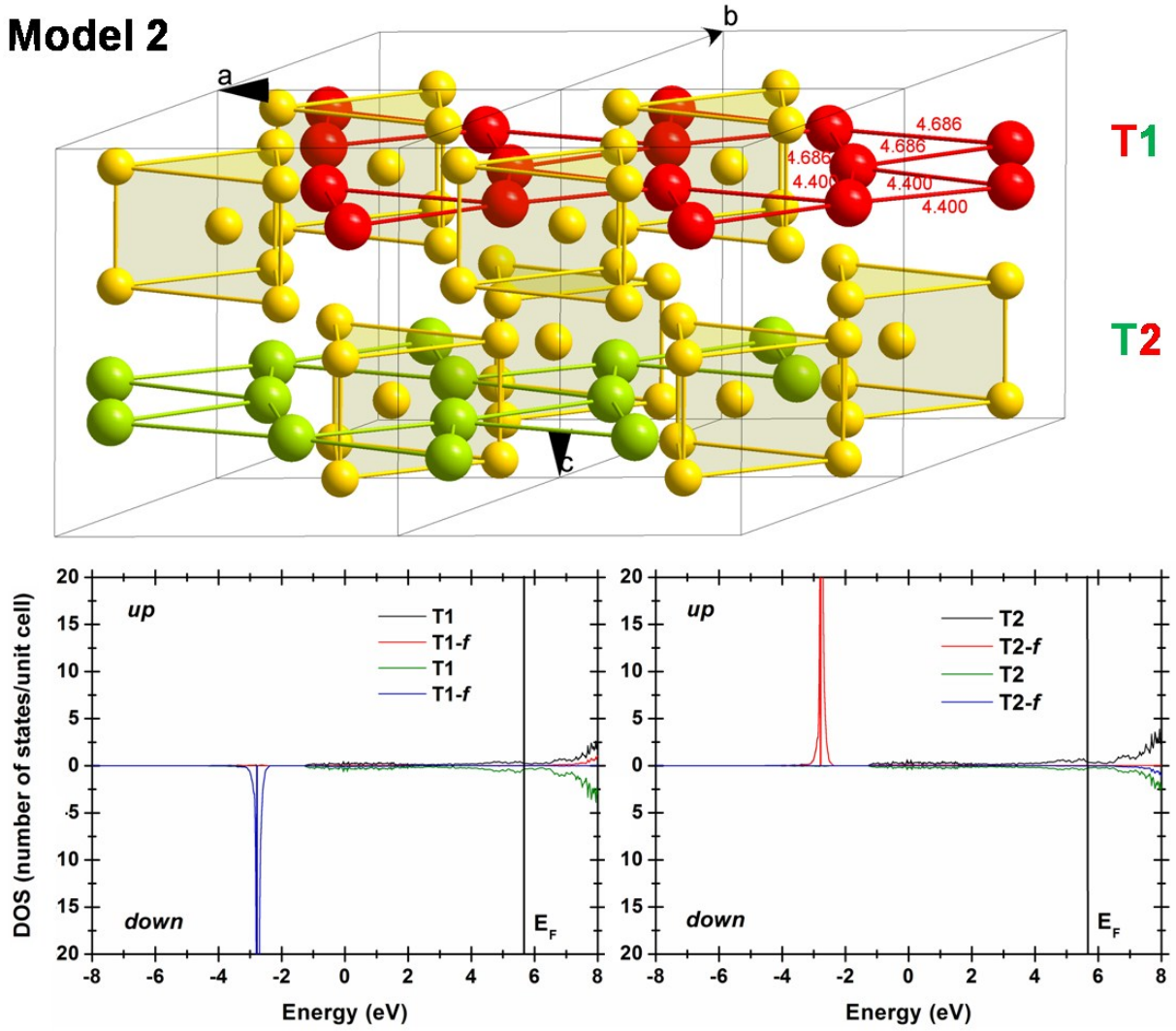

Figure S7. **Top**: Illustration of the antiferromagnetic spin ordering model 2 for the layers of the isosceles Gd triangles (T1 and T2). Spin-up gadolinium atoms are represented in green, while spin-down Gd atoms are highlighted in red. The Au@Au_6 clusters (yellow) are embedded within the Gd nets and the Sn atoms are omitted for clarity. **Bottom**: T1 and T2 layers contribute to the spin-down and spin-up Gd *f* PDOS curves, respectively.

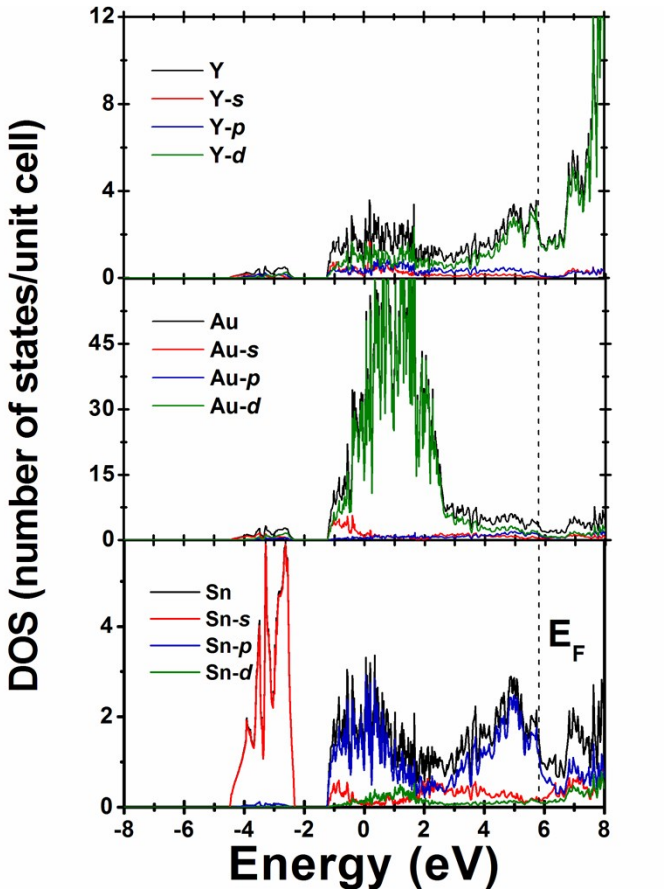

Figure S8. Site and orbital projected DOS curves (VASP) for the hexagonal $Y_3Au_7Sn_3$. The Fermi level is represented by the dashed lines.