Supporting Information

Taking Advantage of Gold's Electronegativity in $R_4Mn_{3-x}Au_{10+x}$ (R = Gd, Y; $0.2 \leq x \leq 1$)

Saroj L. Samal,[1,2] Abhishek Pandey,[1] David C. Johnston,[1,3] John D. Corbett[†1,2]

and Gordon J. Miller*[1,2]

[1]*Ames Laboratory-USDOE* ,[2]*Department of Chemistry, and* [3]*Department of Physics and Astronomy*

Iowa State University, Ames, Iowa 50011, USA

† deceased

* e-mail: gmiller@iastate.edu

Table S1. Loaded (nominal) and refined compositions of $Gd_4Mn_{3-x}Au_{10+x}$ ($0.2 \leq x \leq 1.5$) and the corresponding lattice parameters.

Loaded composition	Refined composition	Lattice parameters (Å)		
		a	*b*	*c*
$Gd_4Mn_{2.8}Au_{10.2}$	$Gd_4Mn_{2.78(1)}Au_{10.22(1)}$	13.581(2)	9.924(2)	10.015(2)
$Gd_4Mn_{2.75}Au_{10.25}$	$Gd_4Mn_{2.74(1)}Au_{10.26(1)}$	13.571(2)	9.929(2)	10.021(2)
$Gd_4Mn_{2.4}Au_{10.6}$	$Gd_4Mn_{2.38(1)}Au_{10.62(1)}$	13.558(2)	9.926(2)	10.052(2)
$Gd_4Mn_2Au_{11}$	$Gd_4Mn_{1.98(2)}Au_{11.02(2)}$	13.501(2)	9.919(2)	10.061(2)
$Gd_4Mn_{1.5}Au_{11.5}$	----	13.454(2)	9.926(2)	10.092(2)

Table S2. Loaded (nominal) and refined compositions of $Y_4Mn_{3-x}Au_{10+x}$ ($0.2 \leq x \leq 0.4$) and the corresponding lattice parameters.

Loaded composition	Refined composition	Lattice parameters (Å)		
		a	*b*	*c*
$Y_4Mn_{2.8}Au_{10.2}$	$Y_4Mn_{2.80(2)}Au_{10.20(2)}$	13.494(2)	9.910(2)	9.964(2)
$Y_4Mn_{2.7}Au_{10.3}$	$Y_4Mn_{2.68(2)}Au_{10.32(2)}$	13.484(2)	9.908(2)	9.971(2)
$Y_4Mn_{2.6}Au_{10.4}$	$Y_4Mn_{2.61(2)}Au_{10.39(2)}$	13.477(2)	9.892(2)	9.981(2)

Table S3. Anisotropic displacement parameters (Å^2) of $Gd_4Mn_{2.78(1)}Au_{10.22(1)}$.

Atom	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Au1	0.0193(4)	0.0177(3)	0.0152(4)	-0.0015(2)	0.0018(3)	-0.0002(3)
Au2	0.0212(4)	0.0123(3)	0.0204(4)	0.0024(2)	0.0021(3)	0.0017(3)
Au3	0.0162(5)	0.0116(4)	0.0142(5)	0.00000	0.00000	0.0010(4)
Mn4	0.018(2)	0.021(1)	0.019 (1)	0.00000	0.0036(12)	0.00000
Au4	0.018(2)	0.021(1)	0.019 (1)	0.00000	0.0036(12)	0.00000
Mn5	0.011(2)	0.010(2)	0.014(2)	0.00000	0.00000	0.0022(19)
Gd1	0.0198(7)	0.0118(5)	0.0146(6)	0.00000	0.00000	0.0014(5)
Gd2	0.0148(6)	0.0128(5)	0.0152(6)	0.00000	0.00000	-0.0007(5)

Table S4. Anisotropic displacement parameters (Å^2) of $Y_4Mn_{2.80(2)}Au_{10.20(2)}$.

Atom	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Au1	0.0255(9)	0.0172(8)	0.0145(7)	-0.0016(6)	0.0021(5)	-0.0007(5)
Au2	0.0179(8)	0.0129(7)	0.0286(8)	0.0013(6)	0.0030(6)	0.0012(5)
Au3	0.015(1)	0.018(1)	0.015(1)	0.00000	0.00000	0.0010(8)
Mn4	0.022(3)	0.028(4)	0.018(3)	0.00000	0.002(2)	0.00000
Au4	0.022(3)	0.028(4)	0.018(3)	0.00000	0.002(2)	0.00000
Mn5	0.012(3)	0.020(3)	0.017(3)	0.00000	0.00000	0.001(4)
Y1	0.022(3)	0.020(2)	0.018(2)	0.00000	0.00000	0.001(2)
Y2	0.013(2)	0.017(2)	0.015(2)	0.00000	0.00000	0.0005(18)

Table S5. All nearest neighbor atom pairs in $Y_4Mn_3Au_{10}$, the ICOHP value for each, and their total contribution per unit cell.

bond	length (Å)	−ICOHP/bond (eV)	No. of bonds/cell (*n*)	−ICOHP/cell (eV)
Y1–Y4	3.862(2)	0.15	8	1.2
Y2 –Y3	3.862(2)	0.15	8	1.2
Y1–Mn2	3.426(2)	0.43	16	6.88
Y3–Mn1	3.460(2)	0.32	16	5.12
Y1–Mn4	3.558(2)	0.30	8	2.4
Y1–Mn3	3.613(2)	0.28	8	2.24
Y3–Mn2	3.712(2)	0.19	16	3.04
Y1–Au5	2.851(1)	1.09	8	8.72
Y3–Au4	2.987(2)	0.65	16	10.4
Y3–Au1	2.994(2)	0.63	16	10.08
Y3–Au5	3.002(2)	0.80	16	12.8
Y3–Au3	3.014(2)	0.74	16	11.84
Y3–Au1	3.029(2)	0.72	16	11.52
Y1–Au2	3.042(2)	0.85	16	13.6
Y1–Au4	3.050(2)	0.85	16	13.6
Y1–Au6	3.104(2)	0.77	8	6.16
Y1–Au6	3.133(2)	0.74	8	5.92
Y1–Au1	3.376(2)	0.48	16	7.68
Y1–Au3	3.394(2)	0.47	16	7.52
Mn1–Au4	2.756(1)	1.26	16	20.16
Mn3–Au4	2.800(1)	1.21	16	19.36
Mn3–Au1	2.820(1)	1.18	16	18.88
Mn1–Au2	2.870(2)	1.13	16	18.08
Mn1–Au1	2.944(2)	1.09	16	17.44
Mn1–Au3	3.100(2)	0.90	16	14.4
Au5–Au5	2.757(1)	1.57	4	6.28
Au3–Au5	2.834(2)	1.38	16	22.08
Au1–Au5	2.837(2)	1.37	16	21.92
Au1–Au4	2.874(2)	1.13	16	18.08
Au1–Au4	2.950(2)	0.96	16	15.36
Au1–Au1	3.041(2)	0.82	8	6.56
Au3–Au4	3.076(2)	0.79	8	6.32

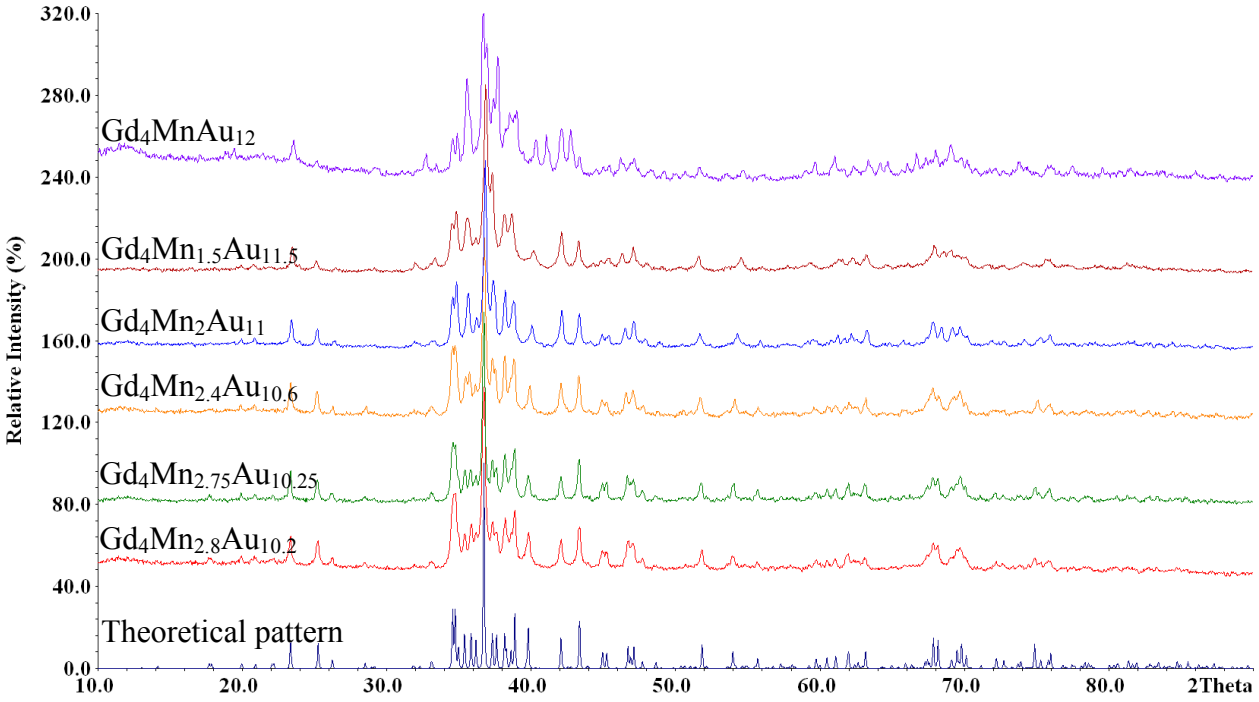

Figure S1. Powder x-ray diffraction patterns of $Gd_4Mn_{3-x}Au_{10+x}$ (x = 0.2, 0.25, 0.6, 1.0, 1.5, 2.0). Bottom line: simulated x-ray diffraction pattern from single crystal data of $Gd_4Mn_{2.78(1)}Au_{10.22(1)}$. Top lines: observed x-ray diffraction patterns.

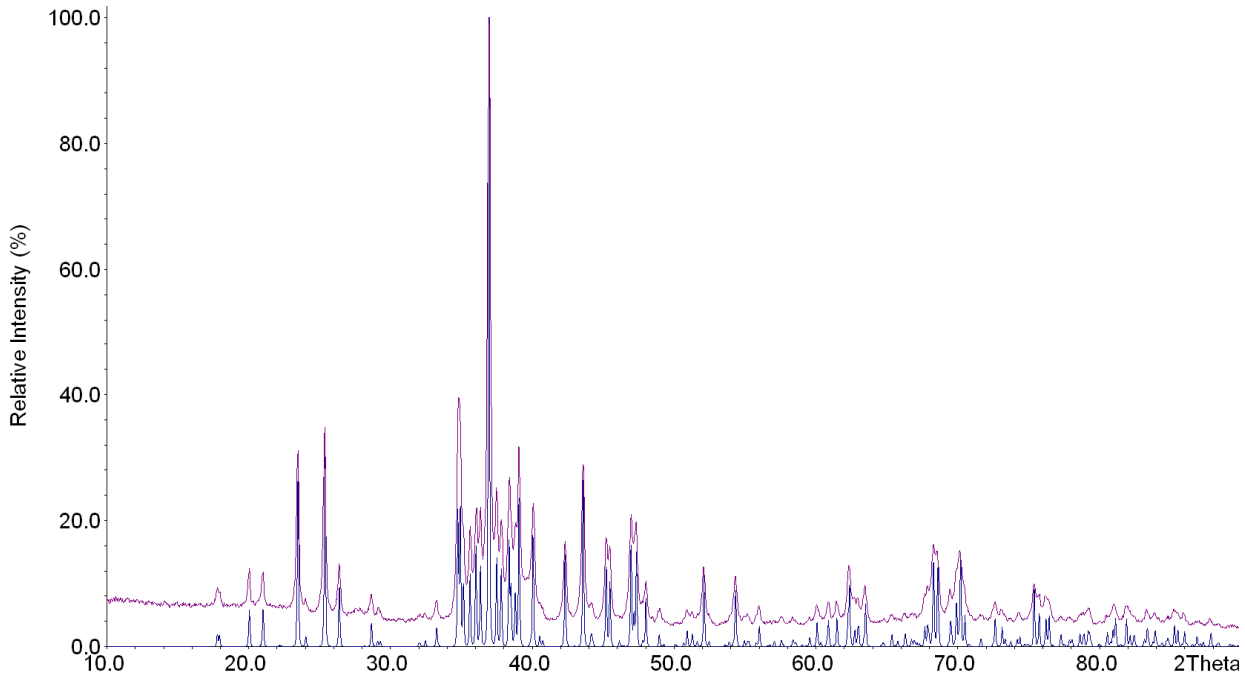

Figure S2. Powder x-ray diffraction pattern of $Y_4Mn_{2.8}Au_{10.2}$. Bottom line: simulated x-ray diffraction pattern from single crystal data of $Y_4Mn_{2.80(2)}Au_{10.20(2)}$. Top line: observed x-ray diffraction pattern.

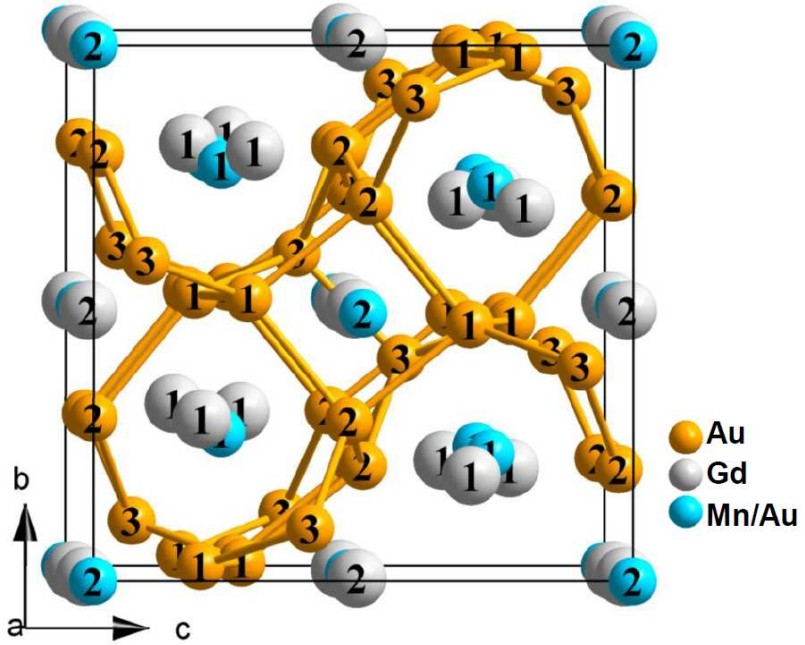

Figure S3. Perspective view of $Gd_4Mn_{2.8}Au_{10.2}$ along the [100] direction showing the sinusoidal layers of Au (yellow Au–Au).

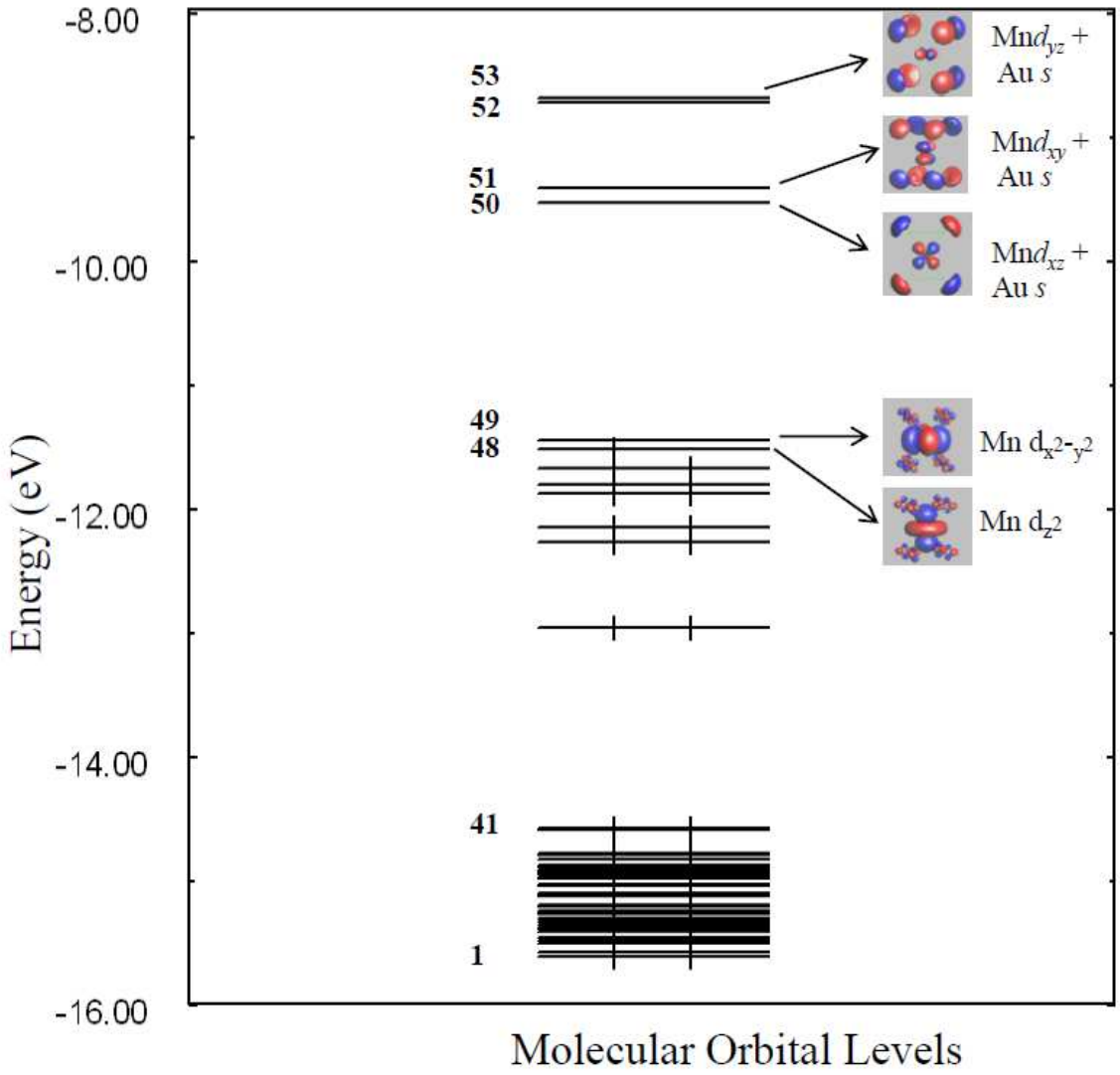

Figure S4. Molecular orbital diagram of Mn@Au_8 molecular species with Mn atom in C_{2h} point symmetry.