

SUPPLEMENTARY DATA

Crystal structure, coloring problem and magnetism of $\text{Gd}_{5-x}\text{Zr}_x\text{Si}_4$

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Table S1. -ICOHP (eV/cell) at the Fermi level for the orthorhombic Gd₅Si₄ and hypothetical Gd₄ZrSi₄ phase (number of equivalent bonds is given in parentheses)

Bonds	Gd ₅ Si ₄		Gd ₄ ZrSi ₄ (O3)	
	Distance (Å)	-ICOHP (E _F)	Distance (Å)	-ICOHP (E _F)
Si1-Si1 (× 4)	2.507(3)	2.344	2.518(2)	2.273
Si2-Si3 (× 4)	2.511(3)	2.367	2.510(2)	2.281
M1-Si1 (× 8)	3.067(2)	1.341	3.013(1)	1.323
-Si1 (× 8)	3.103(2)	1.052	3.065(1)	1.020
-Si1 (× 8)	3.157(2)	1.056	3.113(1)	1.067
-Si1 (× 8)	3.726(2)	0.514	3.602(1)	0.594
-Si2 (× 8)	3.180(2)	0.996	3.131(1)	0.973
-Si3 (× 8)	3.143(2)	1.122	3.077(1)	1.083
-Si3 (× 8)	3.163(2)	0.990	3.109(1)	0.968
M2-Si1 (× 8)	2.887(2)	1.682	2.822(1)	1.673
-Si1 (× 8)	2.905(2)	1.469	2.848(1)	1.473
-Si1 (× 8)	2.995(2)	1.201	2.929(1)	1.203
-Si2 (× 8)	2.972(2)	1.413	2.897(1)	1.384
-Si2 (× 8)	2.977(2)	1.275	2.913(1)	1.265
-Si3 (× 8)	3.058(2)	1.134	2.992(1)	1.099
M3-Si1 (× 8)	3.125(2)	1.059	3.057(1)	1.663
-Si2 (× 4)	2.957(2)	1.388	2.902(1)	2.072
-Si2 (× 4)	3.329(2)	0.774	3.217(1)	1.306
-Si3 (× 4)	2.958(2)	1.352	2.912(1)	2.069
-Si3 (× 4)	3.020(2)	1.272	2.956(1)	1.935
M1-M1 (× 8)	3.8986(3)	0.322	3.8347(3)	0.301
-M1 (× 4)	4.0661(6)	0.466	3.9607(3)	0.450
-M2 (× 8)	3.7751(5)	0.439	3.6816(3)	0.428
-M2 (× 8)	3.8454(5)	0.399	3.7512(3)	0.385
-M2 (× 8)	3.9012(5)	0.395	3.8565(3)	0.360
-M2 (× 8)	4.0587(5)	0.220	3.9851(3)	0.210
-M2 (× 8)	4.0961(5)	0.298	4.0133(3)	0.274
-M3 (× 8)	3.5300(4)	0.389	3.4773(3)	0.642
-M3 (× 8)	3.5856(5)	0.388	3.5171(3)	0.642
M2-M2 (× 4)	3.7793(7)	0.309	3.7043(4)	0.274
-M2 (× 8)	3.9093(5)	0.333	3.8430(3)	0.298
-M3 (× 8)	3.4487(5)	0.493	3.3767(3)	0.728
-M3 (× 8)	3.4499(4)	0.461	3.3777(3)	0.770

Table S2. -ICOHP (eV/cell) at the Fermi level for various models of Zr₅Si₄-type Gd₄ZrSi₄ (number of equivalent bonds is given in parentheses)

Bonds	Distance (Å)	-ICOHP (E _F)		
		Gd ₄ ZrSi ₄ (T1)	Gd ₄ ZrSi ₄ (T2)	Gd ₄ ZrSi ₄ (T3)
Si1-Si2 (× 8)	2.535(9)	2.178	2.151	2.213
M1-Si1 (× 8)	2.989(7)	1.722	1.379	1.407
-Si1 (× 8)	3.107(7)	1.244	0.981	0.986
-Si1 (× 8)	3.150(8)	1.254	0.955	1.021
-Si1 (× 8)	3.559(6)	0.768	0.594	0.640
-Si2 (× 8)	3.064(7)	1.351	1.076	1.053
-Si2 (× 8)	3.078(6)	1.457	1.117	1.087
-Si2 (× 8)	3.222(7)	1.112	0.842	0.845
M2-Si1 (× 8)	2.820(8)	1.608	2.036	1.633
-Si1 (× 8)	2.824(8)	1.429	1.861	1.514
-Si1 (× 8)	2.954(6)	1.153	1.428	1.145
-Si2 (× 8)	2.881(7)	1.422	1.778	1.354
-Si2 (× 8)	2.898(6)	1.276	1.580	1.223
-Si2 (× 8)	2.969(7)	1.143	1.459	1.109
M3-Si1 (× 8)	3.097(8)	0.987	0.977	1.582
-Si2 (× 8)	2.927(6)	1.307	1.296	2.025
-Si2 (× 8)	3.015(7)	1.153	1.132	1.795
M1-M1 (× 8)	3.832(1)	0.498	0.330	0.325
-M1 (× 4)	3.933(2)	0.708	0.418	0.438
-M2 (× 8)	3.688(2)	0.540	0.550	0.431
-M2 (× 8)	3.709(2)	0.515	0.532	0.391
-M2 (× 8)	3.868(2)	0.422	0.429	0.338
-M2 (× 8)	3.969(2)	0.368	0.376	0.280
-M2 (× 8)	4.021(2)	0.237	0.244	0.180
-M2 (× 8)	4.106(2)	0.213	0.219	0.156
-M3 (× 8)	3.544(1)	0.502	0.386	0.632
-M3 (× 8)	3.548(2)	0.486	0.366	0.597
M2-M2 (× 4)	3.754(2)	0.332	0.518	0.320
-M2 (× 8)	3.879(2)	0.264	0.432	0.270
-M3 (× 8)	3.330(2)	0.459	0.588	0.723
-M3 (× 8)	3.348(2)	0.472	0.599	0.742