Supporting Information

Table S1. Selected distances(Å) and angles(°) for $[Ni(pda)_2]Cu_4Sb_2S_6$ (**1**) and $[Ni(dien)_2]CuSb_3S_6$ (**2**)

1			
Sb(1)-S(1)#1	2.435(4)	S(1)#1-Sb(1)-S(3)	99.48(13)
Sb(1)-S(3)	2.436(4)	S(1)#1-Sb(1)-S(2)#2	99.68(13)
Sb(1)-S(2)#2	2.438(4)	S(3)-Sb(1)-S(2)#2	101.10(14)
Cu(1)-S(2)	2.262(4)	S(2)-Cu(1)-S(1)	124.98(15)
Cu(1)-S(1)	2.262(4)	S(2)-Cu(1)-S(3)	120.21(15)
Cu(1)-S(3)	2.274(4)	S(1)-Cu(1)-S(3)	113.00(15)
Cu(2)-S(3)	2.255(4)	S(3)-Cu(2)-S(2)#1	127.36(16)
Cu(2)-S(2)#1	2.273(4)	S(3)-Cu(2)-S(1)#3	117.83(16)
Cu(2)-S(1)#3	2.278(4)	S(2)#1-Cu(2)-S(1)#3	114.49(15)
Ni(1)-N(1)	2.077(11)	N(1)#4-Ni(1)-N(1)	180
Ni(1)-N(2)	2.079(11)	N(2)-Ni(1)-N(2)#4	180
Ni(1)-S(1)	2.633(4)	S(1)-Ni(1)-S(1)#4	180
2			
Sb(1)-S(2)	2.3788(11)	Ni(1)-N(3)	2.138(4)
Sb(1)-S(3)	2.4672(12)	Ni(1)-N(2)	2.146(4)
Sb(1)-S(1)	2.4767(13)	S(2)-Sb(1)-S(3)	100.65(4)
Sb(2)-S(5)	2.3522(11)	S(2)-Sb(1)-S(1)	101.61(5)
Sb(2)-S(3)	2.4607(12)	S(3)-Sb(1)-S(1)	86.57(4)
Sb(2)-S(4)	2.4727(12)	S(5)-Sb(2)-S(3)	99.82(4)
Sb(3)-S(6)	2.3565(12)	S(5)-Sb(2)-S(4)	97.85(4)
Sb(3)-S(1)#1	2.4513(14)	S(3)-Sb(2)-S(4)	95.19(4)
Sb(3)-S(4)#2	2.4661(11)	S(6)-Sb(3)-S(1)#1	101.42(5)
Sb(3)-Cu(1)	3.0873(9)	S(6)-Sb(3)-S(4)#2	101.01(4)
Cu(1)-S(2)#1	2.2531(13)	S(1)#1-Sb(3)-S(4)#2	94.55(4)
Cu(1)-S(5)	2.2553(12)	N(1)-Ni(1)-N(1)#3	180
Cu(1)-S(6)	2.3137(13)	N(3)#3-Ni(1)-N(3)	180
Ni(1)-N(1)	2.087(3)	N(2)-Ni(1)-N(2)#3	180
Ni(1)-N(3)#3	2.138(4)		

Symmetry codes for **1**: #1=x-1,y,z; #2=x-1/2,-y+3/2,z-1/2; #3=x-1/2,-y+3/2,z+1/2 #4=-x+1,-y+1,-z+1; for **2**: #1=x,-y+1/2,z-1/2; #2=x,y+1,z; #3=-x,-y+1,-z+1.

Table S2. Proposed hydrogen bonding geometry for $[Ni(pda)_2]Cu_4Sb_2S_6$ (**1**) and $[Ni(dien)_2]CuSb_3S_6$ (**2**)

D-H	A	d(H···A)	d(D···A)	<DHA	Symmetry code
			1		
N1-H1A	S3	2.60	3.428(12)	153	x+1,y,z
N1-H1B	S2	2.64	3.524(12)	169	x+1/2,-y+1/2,z+1/2
N2-H2A	S3	2.69	3.586(13)	176	x+1/2,-y+1/2,z-1/2
N2-H2B	S2	2.60	3.476(12)	164	-x+1,-y+1,-z+1
			2		
N1-H1C	S6	2.60	3.457(4)	157	
N2-H2B	S6	2.70	3.506(4)	149	
N5-H5C	S2	2.56	3.430(4)	161	-x,-y+1,-z+1