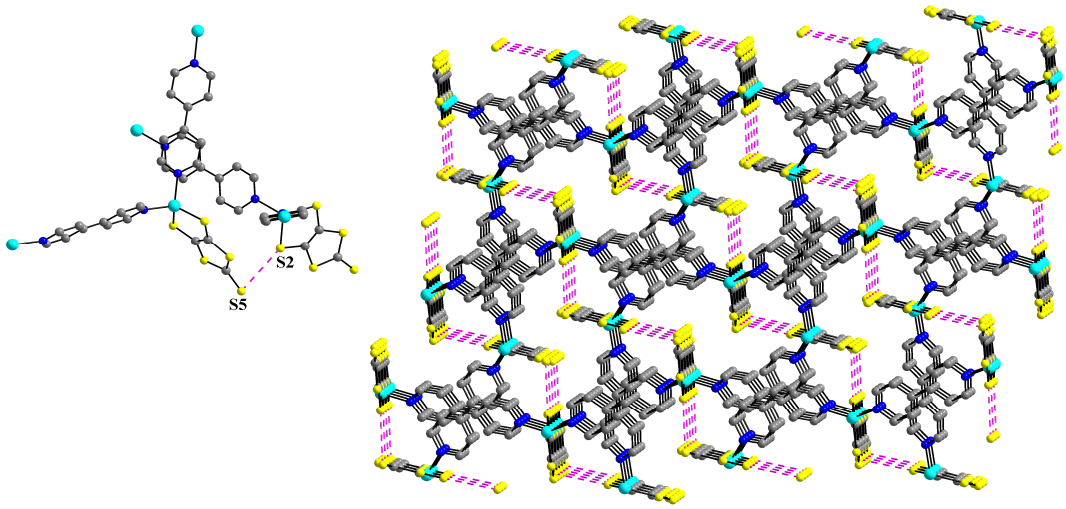
Electronic Supplementary Materials (ESI)

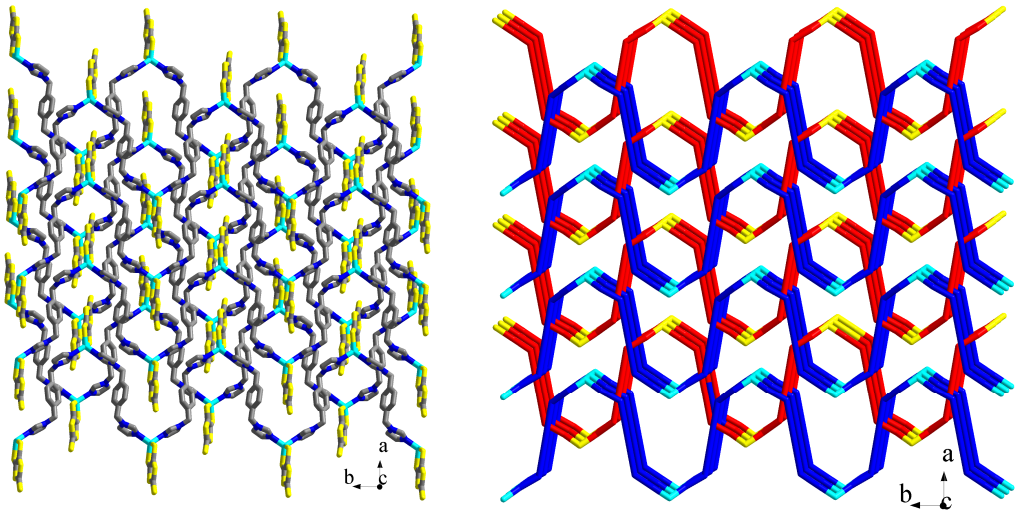
Figure S1



(a) **(b)**

Figure S1. (a) Complementary S···S intermolecular interaction between different directions of perpendicular zig–zag chains of **1**; (b) packing diagram of **1** viewed down the crystallographic a axis in which purple dotted lines indicates the S···S contacts. Color code: Zn, cyan; N, blue; S, yellow; C, gray.

Figure S2



(a) **(b)**

Figure S2. The crystal packing diagram of $[Zn(dmit)(bix)]_n$ (**3**) down the crystallographic *c* axis. (a) View (wire frame representation) and (b) the side view representation of packing diagram. Color code: Zn, cyan; N, blue; S, yellow; C, gray.

Table S1. Bond lengths [Å] and angles [deg] for [**Zn(dmit)(4,4′-bpy)]$_n$** (**compound1**)

Zn(1)-N(1)	2.052(3)	Zn(1)-N(2)	2.063(3)
Zn(1)-S(1)	2.2772(11)	Zn(1)-S(2)	2.3117(11)
S(3)-C(3)	1.724(4)	S(3)-C(2)	1.749(4)
S(1)-C(1)	1.752(4)	S(4)-C(3)	1.723(4)
S(4)-C(1)	1.747(4)	S(2)-C(2)	1.746(4)
S(5)-C(3)	1.660(4)	N(2)-C(9)	1.335(5)
N(2)-C(13)	1.349(5)	C(11)-C(12)	1.400(5)
C(11)-C(10)	1.401(5)	C(11)-C(11)#1	1.479(7)
C(6)-C(7)	1.393(5)	C(6)-C(6)#2	1.488(7)
N(1)-C(8)	1.331(5)	N(1)-C(4)	1.333(5)
C(12)-C(13)	1.389(5)	C(8)-C(7)	1.382(5)
C(10)-C(9)	1.383(5)	C(5)-C(4)	1.381(5)
N(1)-Zn(1)-N(2)	105.19(12)	N(1)-Zn(1)-S(1)	113.01(9)
N(2)-Zn(1)-S(1)	119.15(9)	N(1)-Zn(1)-S(2)	114.97(9)
N(2)-Zn(1)-S(2)	105.78(9)	S(1)-Zn(1)-S(2)	98.87(4)
C(3)-S(3)-C(2)	98.59(18)	C(1)-S(1)-Zn(1)	93.38(13)
C(3)-S(4)-C(1)	98.47(18)	C(2)-S(2)-Zn(1)	92.70(13)
C(9)-N(2)-C(13)	117.9(3)	C(9)-N(2)-Zn(1)	122.6(3)
C(13)-N(2)-Zn(1)	118.7(2)	C(12)-C(11)-C(10)	116.6(3)
C(12)-C(11)-C(11)#1	121.3(4)	C(10)-C(11)-C(11)#1	122.1(4)
C(1)-C(2)-S(2)	127.6(3)	C(1)-C(2)-S(3)	115.2(3)
S(2)-C(2)-S(3)	117.2(2)	C(5)-C(6)-C(7)	116.5(3)
C(5)-C(6)-C(6)#2	121.6(4)	C(7)-C(6)-C(6)#2	121.9(4)
C(2)-C(1)-S(4)	115.7(3)	C(2)-C(1)-S(1)	127.4(3)
S(4)-C(1)-S(1)	117.0(2)	C(8)-N(1)-C(4)	117.6(3)
C(8)-N(1)-Zn(1)	121.9(3)	C(4)-N(1)-Zn(1)	120.4(3)
C(13)-C(12)-C(11)	119.9(4)	N(1)-C(8)-C(7)	122.9(4)
C(9)-C(10)-C(11)	120.0(4)	C(4)-C(5)-C(6)	119.9(4)
S(5)-C(3)-S(4)	123.4(2)	S(5)-C(3)-S(3)	124.5(2)
S(4)-C(3)-S(3)	112.1(2)	N(2)-C(9)-C(10)	123.0(4)
N(1)-C(4)-C(5)	123.1(4)	N(2)-C(13)-C(12)	122.6(4)

Symmetry transformations used to generate equivalent atoms: #1, -x,-y,-z ; #2, -x,-y+1,-z+1

Table S2. Bond lengths [A] and angles [deg] for [**Zn(dmit)(4,4′-bpe)]$_n$** (**Compound 2**).

Zn(1)-N(2)	2.032(10)	Zn(1)-N(1)	2.040(10)
Zn(1)-S(1)	2.271(4)	Zn(1)-S(2)	2.299(4)
Zn(2)-N(4)	2.021(10)	Zn(2)-N(3)	2.051(10)
Zn(2)-S(7)	2.260(4)	Zn(2)-S(6)	2.309(4)
Zn(3)-N(5)	2.037(11)	Zn(3)-N(6)	2.046(11)
Zn(3)-S(11)	2.274(4)	Zn(3)-S(12)	2.314(4)
Zn(4)-N(8)	2.043(10)	Zn(4)-N(7)	2.071(11)
Zn(4)-S(17)	2.268(4)	Zn(4)-S(16)	2.297(4)
S(8)-C(24)	1.701(13)	S(8)-C(22)	1.730(14)
S(2)-C(2)	1.717(13)	S(4)-C(2)	1.728(12)
S(4)-C(3)	1.754(13)	S(9)-C(24)	1.738(12)
S(9)-C(23)	1.777(14)	S(3)-C(3)	1.714(13)
S(3)-C(1)	1.744(13)	S(1)-C(1)	1.730(13)
S(13)-C(32)	1.729(14)	S(13)-C(33)	1.747(14)
S(16)-C(52)	1.745(14)	S(19)-C(54)	1.702(15)
S(19)-C(53)	1.725(13)	S(11)-C(32)	1.751(14)
S(12)-C(31)	1.759(13)	S(6)-C(23)	1.716(14)
S(17)-C(53)	1.712(16)	S(10)-C(24)	1.646(13)
S(18)-C(52)	1.757(14)	S(18)-C(54)	1.759(14)
S(7)-C(22)	1.740(12)	S(14)-C(33)	1.703(13)
S(14)-C(31)	1.722(15)	S(5)-C(3)	1.620(13)
S(15)-C(33)	1.637(12)	S(20)-C(54)	1.616(14)
C(23)-C(22)	1.370(17)	N(1)-C(4)	1.297(15)
N(1)-C(5)	1.335(15)	N(7)-C(45)	1.295(17)
N(7)-C(44)	1.351(16)	N(5)-C(34)	1.314(17)
N(5)-C(35)	1.372(15)	N(2)-C(17)	1.333(14)
N(2)-C(16)	1.339(14)	C(1)-C(2)	1.404(16)
C(50)-C(48)	1.386(15)	C(50)-C(49)	1.395(17)
C(50)-C(51)	1.451(17)	N(3)-C(15)	1.295(14)

N(3)-C(14)	1.334(16)	C(18)-C(17)	1.367(17)
C(18)-C(20)	1.387(16)	C(12)-C(11)	1.380(16)
C(12)-C(15)	1.387(14)	N(6)-C(46)	1.327(15)
N(6)-C(47)	1.341(13)	C(42)-C(41)	1.375(19)
C(42)-C(45)	1.434(18)	N(4)-C(25)	1.274(16)
N(4)-C(26)	1.310(15)	C(8)-C(7)	1.339(16)
C(8)-C(6)	1.394(16)	C(8)-C(9)	1.465(14)
C(52)-C(53)	1.409(18)	C(20)-C(19)	1.406(17)
C(20)-C(21)	1.446(18)	C(32)-C(31)	1.375(17)
C(41)-C(43)	1.39(2)	C(41)-C(40)	1.49(2)
C(27)-C(29)	1.385(18)	C(27)-C(30)	1.391(19)
C(27)-C(28)	1.47(2)	C(58)-C(56)	1.361(17)
C(58)-C(59)	1.399(15)	C(9)-C(10)	1.334(17)
C(43)-C(44)	1.322(18)	C(19)-C(16)	1.383(17)
C(47)-C(49)	1.366(17)	C(14)-C(13)	1.364(14)
C(59)-C(60)	1.420(17)	C(59)-C(57)	1.435(17)
C(11)-C(13)	1.409(17)	C(11)-C(10)	1.474(14)
C(30)-C(26)	1.432(19)	C(56)-N(8)	1.338(15)
C(7)-C(4)	1.404(15)	C(57)-C(55)	1.325(17)
C(5)-C(6)	1.385(14)	C(38)-C(37)	1.32(2)
C(38)-C(36)	1.41(2)	C(38)-C(39)	1.47(2)
C(55)-N(8)	1.346(14)	C(40)-C(39)	1.21(2)
C(37)-C(35)	1.345(18)	C(48)-C(46)	1.353(17)
C(21)-C(28)#1	1.288(18)	C(51)-C(60)#2	1.328(16)
C(60)-C(51)#3	1.328(16)	C(25)-C(29)	1.420(19)
C(28)-C(21)#4	1.287(18)	C(34)-C(36)	1.374(19)
N(2)-Zn(1)-N(1)	97.8(4)	N(2)-Zn(1)-S(1)	113.1(3)
N(1)-Zn(1)-S(1)	118.6(3)	N(2)-Zn(1)-S(2)	122.0(3)
N(1)-Zn(1)-S(2)	107.6(3)	S(1)-Zn(1)-S(2)	99.02(13)
N(4)-Zn(2)-N(3)	100.0(4)	N(4)-Zn(2)-S(7)	117.6(3)
N(3)-Zn(2)-S(7)	115.8(4)	N(4)-Zn(2)-S(6)	116.8(3)
N(3)-Zn(2)-S(6)	108.4(3)	S(7)-Zn(2)-S(6)	98.68(13)
N(5)-Zn(3)-N(6)	98.9(4)	N(5)-Zn(3)-S(11)	118.0(4)
N(6)-Zn(3)-S(11)	114.6(3)	N(5)-Zn(3)-S(12)	108.7(3)
N(6)-Zn(3)-S(12)	118.7(3)	S(11)-Zn(3)-S(12)	98.96(14)

N(8)-Zn(4)-N(7)	100.0(4)	N(8)-Zn(4)-S(17)	109.9(3)
N(7)-Zn(4)-S(17)	117.3(3)	N(8)-Zn(4)-S(16)	122.7(3)
N(7)-Zn(4)-S(16)	108.2(3)	S(17)-Zn(4)-S(16)	99.87(15)
C(24)-S(8)-C(22)	99.2(6)	C(2)-S(2)-Zn(1)	93.1(4)
C(2)-S(4)-C(3)	100.6(6)	C(24)-S(9)-C(23)	98.8(7)
C(3)-S(3)-C(1)	100.7(6)	C(1)-S(1)-Zn(1)	93.5(5)
C(32)-S(13)-C(33)	98.5(6)	C(52)-S(16)-Zn(4)	91.5(5)
C(54)-S(19)-C(53)	101.4(7)	C(32)-S(11)-Zn(3)	93.9(5)
C(31)-S(12)-Zn(3)	93.0(5)	C(23)-S(6)-Zn(2)	91.7(5)
C(53)-S(17)-Zn(4)	94.2(5)	C(52)-S(18)-C(54)	99.1(7)
C(22)-S(7)-Zn(2)	94.5(5)	C(33)-S(14)-C(31)	99.8(7)
C(22)-C(23)-S(6)	130.2(11)	C(22)-C(23)-S(9)	112.8(10)
S(6)-C(23)-S(9)	116.9(8)	C(4)-N(1)-C(5)	116.3(11)
C(4)-N(1)-Zn(1)	120.8(9)	C(5)-N(1)-Zn(1)	122.9(9)
C(45)-N(7)-C(44)	117.3(12)	C(45)-N(7)-Zn(4)	120.0(9)
C(44)-N(7)-Zn(4)	122.6(10)	C(34)-N(5)-C(35)	117.7(12)
C(34)-N(5)-Zn(3)	122.7(10)	C(35)-N(5)-Zn(3)	119.6(10)
C(17)-N(2)-C(16)	112.9(10)	C(17)-N(2)-Zn(1)	123.5(8)
C(16)-N(2)-Zn(1)	123.6(8)	C(2)-C(1)-S(1)	126.7(10)
C(2)-C(1)-S(3)	114.6(9)	S(1)-C(1)-S(3)	118.7(8)
C(48)-C(50)-C(49)	114.7(13)	C(48)-C(50)-C(51)	122.3(12)
C(49)-C(50)-C(51)	122.8(12)	C(15)-N(3)-C(14)	116.0(11)
C(15)-N(3)-Zn(2)	121.8(9)	C(14)-N(3)-Zn(2)	122.1(9)
C(17)-C(18)-C(20)	119.2(12)	C(23)-C(22)-S(8)	117.2(9)
C(23)-C(22)-S(7)	124.5(11)	S(8)-C(22)-S(7)	118.3(8)
C(11)-C(12)-C(15)	116.9(11)	C(46)-N(6)-C(47)	116.3(11)
C(46)-N(6)-Zn(3)	123.4(8)	C(47)-N(6)-Zn(3)	120.2(9)
C(41)-C(42)-C(45)	117.1(15)	C(25)-N(4)-C(26)	117.0(12)
C(25)-N(4)-Zn(2)	120.9(9)	C(26)-N(4)-Zn(2)	121.8(9)
C(7)-C(8)-C(6)	117.8(10)	C(7)-C(8)-C(9)	124.5(11)
C(6)-C(8)-C(9)	117.7(11)	C(53)-C(52)-S(16)	128.4(11)
C(53)-C(52)-S(18)	114.1(11)	S(16)-C(52)-S(18)	117.2(9)
C(18)-C(20)-C(19)	115.3(12)	C(18)-C(20)-C(21)	120.0(13)
C(19)-C(20)-C(21)	124.5(12)	C(31)-C(32)-S(13)	115.3(10)
C(31)-C(32)-S(11)	127.2(11)	S(13)-C(32)-S(11)	117.5(8)

S(15)-C(33)-S(14)	125.9(9)	S(15)-C(33)-S(13)	122.9(8)
S(14)-C(33)-S(13)	111.1(6)	C(42)-C(41)-C(43)	117.8(14)
C(42)-C(41)-C(40)	123.2(18)	C(43)-C(41)-C(40)	118.5(15)
C(32)-C(31)-S(14)	115.0(10)	C(32)-C(31)-S(12)	126.8(11)
S(14)-C(31)-S(12)	117.9(8)	C(29)-C(27)-C(30)	118.7(14)
C(29)-C(27)-C(28)	120.2(14)	C(30)-C(27)-C(28)	121.1(14)
C(56)-C(58)-C(59)	120.5(13)	C(10)-C(9)-C(8)	123.3(12)
C(44)-C(43)-C(41)	120.6(14)	C(16)-C(19)-C(20)	119.8(12)
N(6)-C(47)-C(49)	123.6(12)	S(5)-C(3)-S(3)	126.4(8)
S(5)-C(3)-S(4)	123.7(8)	S(3)-C(3)-S(4)	109.9(7)
C(52)-C(53)-S(17)	125.4(10)	C(52)-C(53)-S(19)	114.6(12)
S(17)-C(53)-S(19)	119.8(8)	N(3)-C(14)-C(13)	124.5(12)
C(58)-C(59)-C(60)	118.7(12)	C(58)-C(59)-C(57)	113.8(12)
C(60)-C(59)-C(57)	127.5(11)	C(12)-C(11)-C(13)	118.3(10)
C(12)-C(11)-C(10)	119.4(11)	C(13)-C(11)-C(10)	122.3(12)
S(10)-C(24)-S(8)	125.4(7)	S(10)-C(24)-S(9)	122.8(8)
S(8)-C(24)-S(9)	111.9(7)	N(7)-C(45)-C(42)	123.4(13)
C(9)-C(10)-C(11)	127.4(12)	C(27)-C(30)-C(26)	118.1(13)
N(2)-C(17)-C(18)	127.3(11)	N(8)-C(56)-C(58)	124.9(12)
N(2)-C(16)-C(19)	125.2(12)	N(3)-C(15)-C(12)	126.3(12)
N(1)-C(5)-C(6)	124.7(12)	C(37)-C(38)-C(36)	117.3(14)
C(37)-C(38)-C(39)	118.4(16)	C(36)-C(38)-C(39)	123.9(17)
C(57)-C(55)-N(8)	125.5(13)	C(1)-C(2)-S(2)	127.2(9)
C(1)-C(2)-S(4)	114.1(10)	S(2)-C(2)-S(4)	118.6(8)
C(47)-C(49)-C(50)	120.2(12)	C(5)-C(6)-C(8)	117.6(11)
C(39)-C(40)-C(41)	130(2)	C(56)-N(8)-C(55)	114.6(11)
C(56)-N(8)-Zn(4)	124.5(9)	C(55)-N(8)-Zn(4)	121.0(9)
S(20)-C(54)-S(19)	126.0(9)	S(20)-C(54)-S(18)	123.4(9)
S(19)-C(54)-S(18)	110.5(7)	C(14)-C(13)-C(11)	117.9(12)
N(4)-C(26)-C(30)	122.9(14)	C(38)-C(37)-C(35)	121.4(15)
C(43)-C(44)-N(7)	123.6(15)	C(46)-C(48)-C(50)	121.8(13)
C(28)#1-C(21)-C(20)	127.1(15)	C(40)-C(39)-C(38)	129(2)
C(60)#2-C(51)-C(50)	126.6(12)	C(37)-C(35)-N(5)	122.1(14)
C(51)#3-C(60)-C(59)	125.6(13)	N(4)-C(25)-C(29)	127.4(13)

C(21)#4-C(28)-C(27)	127.2(15)	C(27)-C(29)-C(25)	115.7(13)
N(6)-C(46)-C(48)	123.2(12)	N(5)-C(34)-C(36)	121.6(14)

Symmetry transformations used to generate equivalent atoms: #1 x,y,z-1; #2 x,y-1,z; #3 x,y+1,z; #4 x,y,z+1

Table S3. Bond lengths [A] and angles [deg] for [**Zn(dmit)(bix)]$_n$ (Compound 3)**

Zn(1)-N(3)	2.014(3)	Zn(1)-N(1)	2.021(3)
Zn(1)-S(2)	2.2963(11)	Zn(1)-S(1)	2.3329(11)
S(3)-C(3)	1.714(4)	S(3)-C(1)	1.747(3)
S(1)-C(1)	1.743(4)	S(2)-C(2)	1.748(4)
S(4)-C(3)	1.716(4)	S(4)-C(2)	1.746(4)
S(5)-C(3)	1.662(4)	C(1)-C(2)	1.343(5)
C(8)-C(10)	1.358(5)	C(8)-C(9)	1.358(5)
C(8)-C(7)	1.501(5)	N(3)-C(12)	1.316(4)
N(3)-C(11)	1.355(5)	N(1)-C(4)	1.307(4)
N(1)-C(5)	1.351(5)	C(15)-C(17)	1.353(5)
C(15)-C(16)	1.366(5)	C(15)-C(14)	1.513(5)
C(4)-N(2)	1.325(4)	N(4)-C(12)	1.340(4)
N(4)-C(13)	1.354(5)	N(4)-C(14)	1.466(4)
N(2)-C(6)	1.353(5)	N(2)-C(7)	1.471(4)
C(13)-C(11)	1.350(5)	C(16)-C(10)#1	1.373(6)
C(9)-C(17)#2	1.378(5)	C(10)-C(16)#2	1.373(6)
C(5)-C(6)	1.343(5)	C(17)-C(9)#1	1.378(5)
N(3)-Zn(1)-N(1)	108.28(12)	N(3)-Zn(1)-S(2)	116.45(9)
N(1)-Zn(1)-S(2)	119.28(9)	N(3)-Zn(1)-S(1)	107.51(10)
N(1)-Zn(1)-S(1)	107.17(9)	S(2)-Zn(1)-S(1)	96.50(4)
C(3)-S(3)-C(1)	98.86(18)	C(1)-S(1)-Zn(1)	93.81(12)
C(2)-S(2)-Zn(1)	95.21(13)	C(3)-S(4)-C(2)	98.41(19)
C(2)-C(1)-S(1)	128.1(3)	C(2)-C(1)-S(3)	114.9(3)
S(1)-C(1)-S(3)	116.9(2)	C(10)-C(8)-C(9)	117.2(4)
C(10)-C(8)-C(7)	120.7(4)	C(9)-C(8)-C(7)	122.1(4)
C(1)-C(2)-S(4)	115.9(3)	C(1)-C(2)-S(2)	126.3(3)

S(4)-C(2)-S(2)	117.8(2)	C(12)-N(3)-C(11)	106.0(3)
C(12)-N(3)-Zn(1)	121.8(3)	C(11)-N(3)-Zn(1)	131.3(3)
C(4)-N(1)-C(5)	104.9(3)	C(4)-N(1)-Zn(1)	122.9(3)
C(5)-N(1)-Zn(1)	130.8(3)	C(17)-C(15)-C(16)	118.1(4)
C(17)-C(15)-C(14)	121.4(4)	C(16)-C(15)-C(14)	120.5(4)
N(1)-C(4)-N(2)	112.8(3)	C(12)-N(4)-C(13)	106.8(3)
C(12)-N(4)-C(14)	125.4(4)	C(13)-N(4)-C(14)	127.8(3)
N(3)-C(12)-N(4)	111.1(3)	C(4)-N(2)-C(6)	105.6(3)
C(4)-N(2)-C(7)	127.4(4)	C(6)-N(2)-C(7)	127.0(3)
N(2)-C(7)-C(8)	112.0(3)	S(5)-C(3)-S(3)	123.0(2)
S(5)-C(3)-S(4)	125.1(3)	S(3)-C(3)-S(4)	111.9(2)
C(11)-C(13)-N(4)	106.9(4)	C(15)-C(16)-C(10)#1	120.9(4)
C(13)-C(11)-N(3)	109.3(4)	C(8)-C(9)-C(17)#2	121.9(4)
C(8)-C(10)-C(16)#2	121.4(4)	C(6)-C(5)-N(1)	109.4(4)
C(15)-C(17)-C(9)#1	120.4(4)	C(5)-C(6)-N(2)	107.3(4)
N(4)-C(14)-C(15)	111.7(3)		

Symmetry transformations used to generate equivalent atoms:#1, -x+1/2,y-1/2,-z+1/2; #2, -x+1/2,y+1/2,-z+1/2