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Supporting Information

Magnetism, dielectrics and ion conduction in two polymorphs of a heteroleptic bis(dithiolato)nickelate molecular solid

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Table S1: Bond lengths and bond angles in both anions and cations of **1α** and **1β** at 293 K.

		1α	
Ni(1)-S(4)	2.1615(8)	Ni(1)-S(5)	2.1661(8)
Ni(1)-S(6)	2.1512(8)	Ni(1)-S(7)	2.1321(8)
S(1)-C(3)	1.731(3)	S(1)-C(2)	1.738(3)
S(2)-C(3)	1.728(3)	S(2)-C(1)	1.736(3)
S(3)-C(3)	1.639(3)	S(4)-C(2)	1.709(3)
S(5)-C(1)	1.697(3)	S(6)-C(4)	1.727(3)
S(7)-C(5)	1.722(3)	N(1)-C(7)	1.131(4)
N(2)-C(6)	1.125(4)	N(3)-C(10)	1.499(3)
N(3)-C(15)	1.506(3)	N(3)-C(8)	1.519(3)
N(3)-C(13)	1.522(4)	C(1)-C(2)	1.364(4)
C(4)-C(5)	1.351(4)	C(4)-C(7)	1.443(4)
C(5)-C(6)	1.430(4)	C(8)-C(9)	1.496(4)
C(10)-C(11)	1.484(4)	C(11)-C(12)	1.484(5)
C(13)-C(14)	1.503(4)	C(15)-C(16)	1.513(4)
S(7)-Ni(1)-S(6)	92.47(3)	S(7)-Ni(1)-S(4)	178.79(3)
S(6)-Ni(1)-S(4)	88.70(3)	S(7)-Ni(1)-S(5)	85.96(3)
S(6)-Ni(1)-S(5)	177.78(3)	S(4)-Ni(1)-S(5)	92.87(3)
		1β	
Ni(1)-S(2)	2.1446(15)	Ni(1)-S(1)	2.1537(16)
Ni(1)-S(4)	2.1542(16)	Ni(1)-S(3)	2.1654(15)
S(1)-C(2)	1.737(6)	S(2)-C(3)	1.708(7)
S(3)-C(5)	1.715(5)	S(4)-C(6)	1.708(5)
S(5)-C(5)	1.730(5)	S(5)-C(7)	1.757(6)
S(6)-C(7)	1.725(6)	S(6)-C(6)	1.726(5)
S(7)-C(7)	1.609(7)	N(1)-C(1)	1.155(9)
N(2)-C(4)	1.114(8)	C(1)-C(2)	1.399(9)
C(2)-C(3)	1.369(10)	C(3)-C(4)	1.446(8)
C(5)-C(6)	1.362(8)	N(3)-C(15)	1.503(5)
N(3)-C(11)	1.511(5)	N(3)-C(8)	1.515(5)
N(3)-C(13)	1.516(5)	C(8)-C(9)	1.508(7)
C(9)-C(10)	1.489(7)	C(11)-C(12)	1.512(7)
C(13)-C(14)	1.508(7)	C(15)-C(16)	1.493(7)
S(2)-Ni(1)-S(1)	92.51(7)	S(2)-Ni(1)-S(4)	86.56(5)
S(1)-Ni(1)-S(4)	179.06(7)	S(2)-Ni(1)-S(3)	179.34(7)
S(1)-Ni(1)-S(3)	88.14(4)	S(4)-Ni(1)-S(3)	92.78(7)

Table S2: Bond lengths (Å) in both anions and cations of **1α** and **1β** after crystal structure optimization.

		1α	
Ni(1)-S(4)	2.229	Ni(1)-S(5)	2.250
Ni(1)-S(6)	2.199	Ni(1)-S(7)	2.192
S(1)-C(3)	1.743	S(1)-C(2)	1.743
S(2)-C(3)	1.745	S(2)-C(1)	1.734
S(3)-C(3)	1.651	S(4)-C(2)	1.706
S(5)-C(1)	1.705	S(6)-C(4)	1.727
S(7)-C(5)	1.717	N(1)-C(7)	1.172
N(2)-C(6)	1.171	N(3)-C(10)	1.524
N(3)-C(15)	1.522	N(3)-C(8)	1.525
N(3)-C(13)	1.526	C(1)-C(2)	1.395
C(4)-C(5)	1.394	C(4)-C(7)	1.411
C(5)-C(6)	1.411	C(8)-C(9)	1.516
C(10)-C(11)	1.520	C(11)-C(12)	1.524
C(13)-C(14)	1.517	C(15)-C(16)	1.518
		1β	
Ni(1)-S(2)	2.210	Ni(1)-S(1)	2.224
Ni(1)-S(4)	2.294	Ni(1)-S(3)	2.257
S(1)-C(2)	1.719	S(2)-C(3)	1.719
S(3)-C(5)	1.720	S(4)-C(6)	1.720
S(5)-C(5)	1.748	S(5)-C(7)	1.745
S(6)-C(7)	1.735	S(6)-C(6)	1.738
S(7)-C(7)	1.654	N(1)-C(1)	1.172
N(2)-C(4)	1.171	C(1)-C(2)	1.414
C(2)-C(3)	1.394	C(3)-C(4)	1.411
C(5)-C(6)	1.386	N(3)-C(15)	1.523
N(3)-C(11)	1.524	N(3)-C(8)	1.529
N(3)-C(13)	1.529	C(8)-C(9)	1.522
C(9)-C(10)	1.527	C(11)-C(12)	1.513
C(13)-C(14)	1.515	C(15)-C(16)	1.515

Table S3: Typical interatomic and plane-to-plane distances (Å) in an anion stack in **1α** and **1β**.

	1α		**1β**
$d_{Ni1...Ni1}$	3.948	$d_{Ni1...Ni1}$	4.200
$d_{plane-plane}$	3.569	$d_{plane-plane}$	3.557
$d_{S2...S2}$	3.391	$d_{N2...H13B}$	2.5728(76)
$d_{S2\#1...S2\#2}$	3.3910 (12)	$d_{N2...H15B}$	2.6208(71)
$d_{S7...H13B}$	2.9854(8)	$d_{S3...H14A}$	2.9829(14)
$d_{N1...H10A}$	2.6410(34)	$d_{S6...H12B}$	2.9337(16)
$d_{N2...H8B}$	2.4048(40)		
#1 = 2-x, -y, 1-z;	#2 = 1+x, -1+y, 1+z		

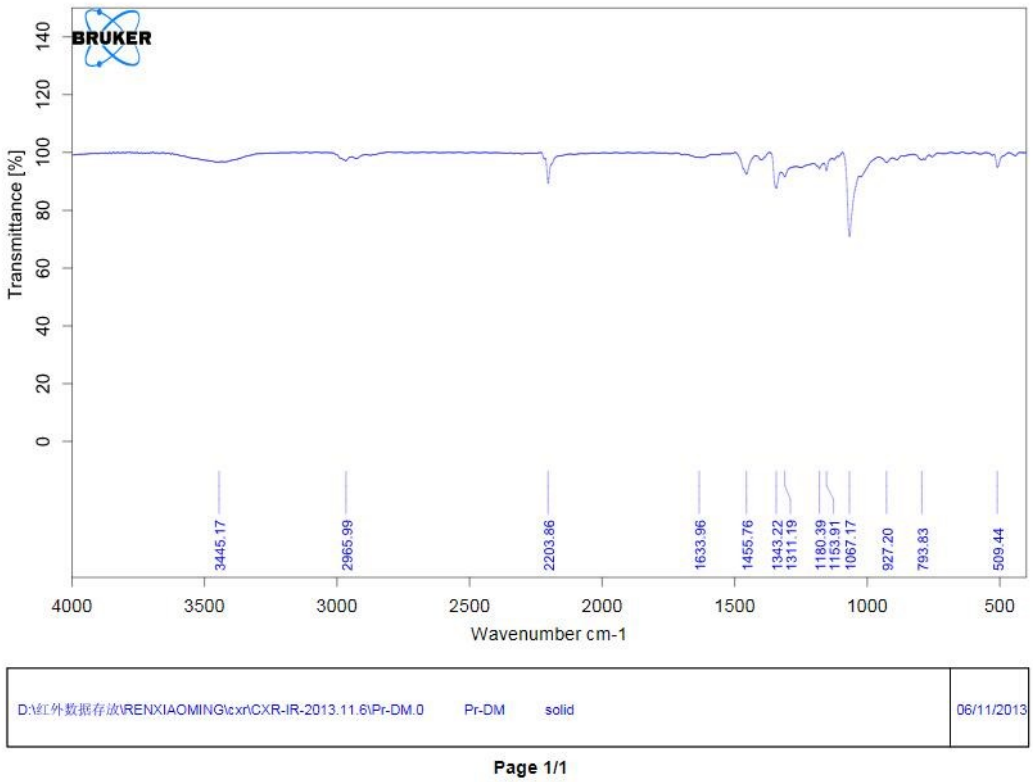

Fig.S1 FT-IR spectrum of **1α**.

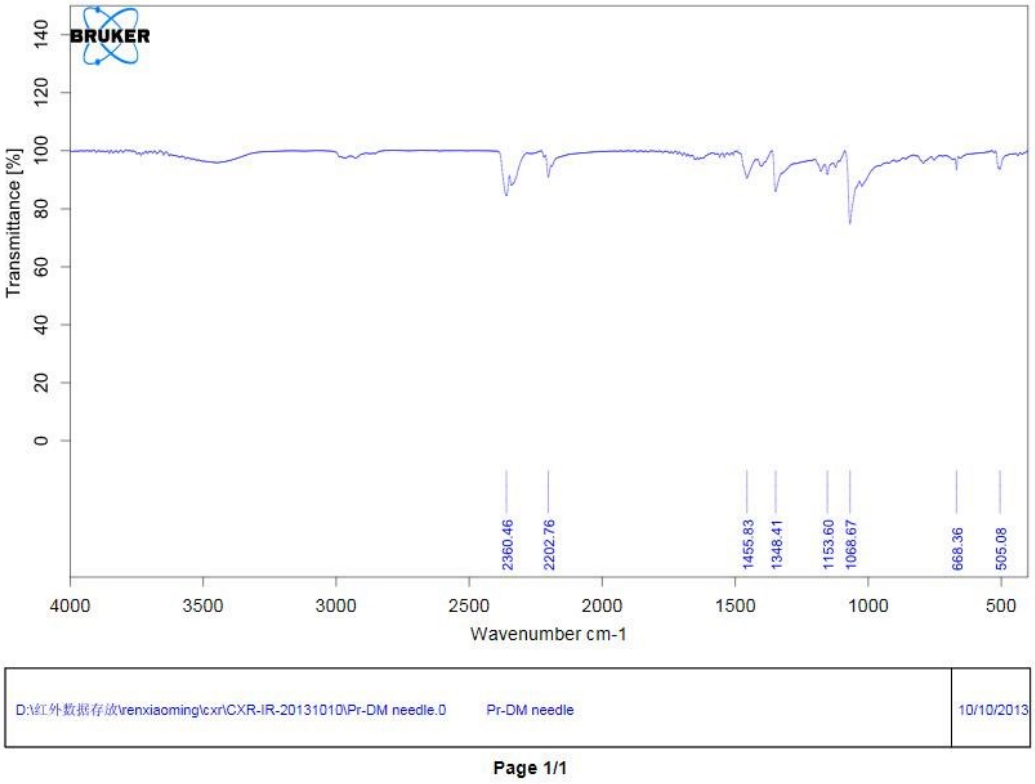

Fig.S2 FT-IR spectrum of **1β**.

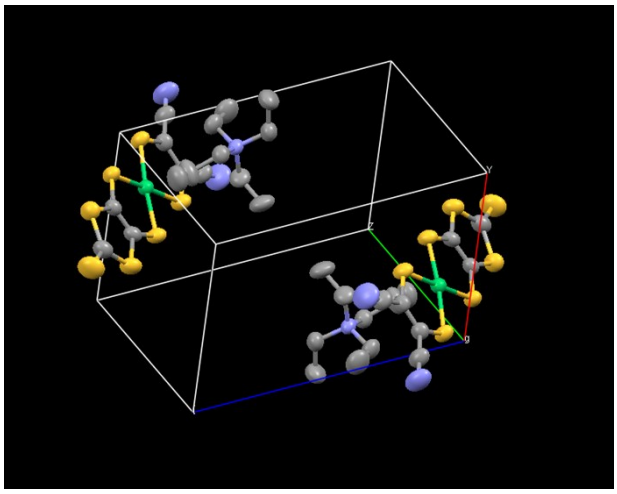

Fig.S3 The optimized crystal structure of **1α** through CASTEP.

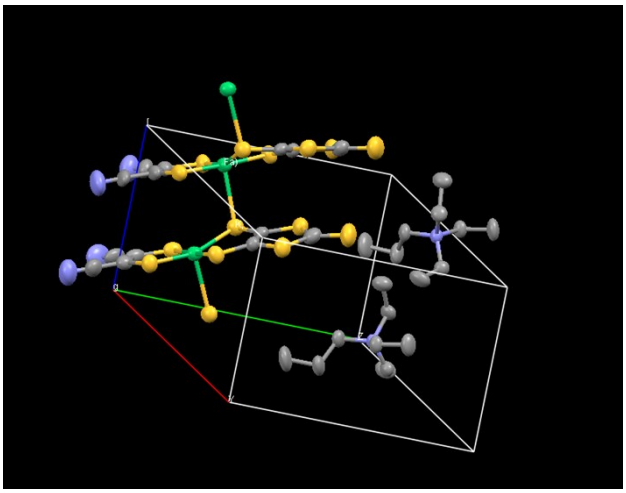

Fig.S4 The optimized crystal structure of **1β** through CASTEP.

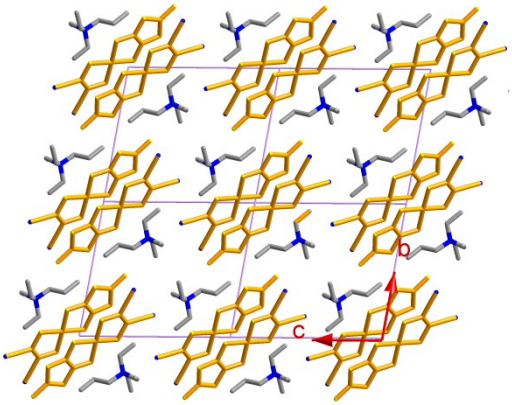

Fig.S5The alternating layers of Et_3PrN^+ cations and [Ni(dmit)(mnt)]- anions of **1α** viewed in the a-axis direction.