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Supporting information for article:

Bis- (1-(2-Aminoethyl)piperidino), (2-(2-aminoethyl)pyridino) and (1-(2-aminoethyl)pyrrolidino)-substituted dicyanoquinodimethanes: consequences of flexible ethylene spacers with heterocyclic moieties and amine functionalities

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S1: Instrumentation Details

Table S1. Crystallographic data for Compound **AEPiDQ**

Table S2. Crystallographic data for Compound **AEPyDQ**

Table S3. Crystallographic data for Compound **AEPrDQ**

Fig. S1 Absorption and the emission spectra of **(a)** AEPiDQ (------, magenta) **(b)** AEPyDQ (……., green) and **(c)** AEPrDQ (-.-.-., grey) respectively.

Table S4: (i) Absorption and emission λ_{max} for AEPiDQ, AEPyDQ and AEPrDQ in acetonitrile.

Table S5: Powder SHG data showing phase-matchable behavior in *Bis*- (2-(2-Amino ethyl)pyridino) dicyanoquinodimethane (AEPyDQ)

Fig. S2 Comparison of powder X-ray data with the single X-ray crystal structure CIF files: PIP, PYP and PYR refers to AEPiDQ, AEPyDQ and AEPrDQ.

Fig. S3 Supramolecular structures and molecular packing view, with arrangement of dipoles in a distinct fashion in (a) AEPiDQ (b) AEPyDQ and AEPrDQ. Hydrogen atoms are omitted for clarity.

Fig. S4 Yellow to pale yellow color observed under ambient light (a), (b), (c) and bright green emission noted under fluorescence microscope images acquired with a 10X objective lens (d), (e) and (f) respectively for AEPiDQ, AEPyDQ and AEPrDQ.

S1. Instrumentation Details

(i) Absorption spectra were studied on data recorded on the Jasco V-650 make and Jasco-ISV-727 model. Fluorescence studies were performed on Horiba JobinYvon, Model FL3-22 Fluorolog spectrofluorimeter. Infrared (IR, 400 - 4000 cm^{-1}) spectra were carried out on JASCO-FTIR-4200 spectrophotometer with samples as KBr pellets.

(ii) Agilent Xcalibur Gemini Diffractometer equipped with EOS CCD detector at 298 K, using graphite-monochromated Mo-Kα radiation (0.7107 Å), was used for data collection. For data collection and data reduction CrysAlisPro, Agilent Technologies, Version 1.171.35.19 program was used. Using spherical harmonics empirical absorption correction was implemented in SCALE3 ABSPACK scaling algorithm. Using Olex 2[37], with direct methods using SHELXS-97[38] the crystal structure was solved and refinement was done against F2 using SHELXL-97[38] refinement package with least square minimization. All non-hydrogen atoms were refined using anisotropic displacement parameters.

(iii) Powder X-ray diffraction data were recorded on SMART Bruker D8 Advance X-ray diffractometer using CuKα radiations with λ = 1.5406 A° at 40 kV and 30 mA over the 2θ range 5-80° at the scan rate of 6°/min. Powder cell program was used to invigorate the diffraction pattern using single crystal structure details.

(iv) Images were captured using inverted fluorescence microscope, DMi 8, Leica microsystems and CCD camera. All images were acquired with a 10X objective lens.

Table S1 Table 1. Crystallographic data for Compound AEPiDQ

Table 1S*a*. Crystal data and structure refinement for AEPiDQ.

Identification code	AEPiDQ
Empirical formula	$C_{24}H_{34}N_6$
Formula weight	406.57
Temperature/K	293(2)
Crystal system	triclinic
Space group	P-1
a/Å	8.7977(9)
b/Å	10.3488(10)
c/Å	13.4887(10)
α/°	84.101(7)
β/°	87.035(7)
γ/°	73.903(9)
Volume/Å^3	1173.34(19)
Z	2
ρ_{calc}mg/mm^3	1.151
m/mm^{-1}	0.071
F(000)	440.0
Crystal size/mm^3	0.42 × 0.34 × 0.32
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection	5.76 to 52.742°
Index ranges	$-10 \le h \le 10$, $-12 \le k \le 11$, $-16 \le l \le 16$
Reflections collected	8002
Independent reflections	4780 [R_{int} = 0.0251, R_{sigma} = 0.0543]
Data/restraints/parameters	4780/0/271
Goodness-of-fit on F^2	1.015
Final R indexes [I>=2σ (I)]	R_1 = 0.0594, wR_2 = 0.1274
Final R indexes [all data]	R_1 = 0.1078, wR_2 = 0.1581
Largest diff. peak/hole / e Å^{-3}	0.25/-0.16

Table S1*b*. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for AEPiDQ. U_{eq} is defined as one third of the trace of the orthogonalized U_{IJ} tensor.

Atom	x	y	z	U(eq)
C1	-374(2)	10484(2)	2226.5(13)	42.9(5)
C2	572(2)	10010(2)	1398.1(14)	45.4(5)
C3	1588(2)	8745(2)	1426.7(13)	45.0(5)
C4	1751(2)	7862(2)	2296.5(13)	44.9(5)
C5	834(3)	8332(2)	3127.6(14)	52.0(6)
C6	-187(3)	9599(2)	3095.6(14)	51.1(5)
C7	-1462(2)	11801(2)	2178.9(14)	48.6(5)
C8	-2319(3)	12327(2)	3027.9(17)	58.2(6)
C9	-1675(3)	12633(2)	1284.2(17)	59.2(6)
C10	2735(2)	6475(2)	2315.5(14)	46.5(5)
C11	4196(3)	6444(2)	3823.1(14)	62.8(7)
C12	5904(3)	6281(2)	3537.1(16)	68.3(7)
C13	5784(3)	8314(3)	2441.5(19)	78.7(8)
C14	5851(4)	8875(3)	1377(2)	94.8(9)
C15	7426(4)	8250(4)	881(2)	97.9(10)
C16	7808(4)	6736(4)	1037(2)	106.1(11)
C17	7683(3)	6253(3)	2125(2)	86.7(9)
C18	3939(3)	4551(2)	1328.2(15)	59.9(6)
C19	3369(3)	3329(2)	1640.3(15)	67.6(7)
C20	1396(3)	3632(2)	2970.6(17)	65.2(6)
C21	1108(3)	3539(3)	4086.8(18)	79.8(8)
C22	1729(4)	2115(3)	4530.1(19)	86.5(9)
C23	3432(4)	1543(3)	4215.2(17)	81.8(8)
C24	3668(3)	1724(2)	3094.6(16)	66.2(7)
N1	-2989(3)	12740(2)	3730.0(17)	89.7(8)
N2	-1808(3)	13275(2)	532.3(15)	89.5(8)
N3	3458(2)	5821.1(17)	3130.0(11)	53.5(5)
N4	6107(2)	6857.5(18)	2527.0(13)	56.5(5)
N5	2814(2)	5833.7(17)	1509.0(11)	51.7(5)

N6	3069(2)	3141.5(16)	2718.7(11)	48.9(4)

Table S1*c*. Anisotropic Displacement Parameters ($Å^2×10^3$) for AEPiDQ. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	U11	U22	U33	U23	U13	U12
C1	45.3(13)	42.3(12)	45.0(11)	-5.6(9)	2.0(9)	-18.4(10)
C2	53.9(13)	42.7(12)	40.1(10)	-0.6(9)	1.2(9)	-15.9(10)
C3	50.6(13)	44.3(12)	40.1(10)	-4.3(9)	6.2(9)	-14.3(10)
C4	50.4(13)	41.5(12)	41.6(11)	0.2(9)	3.7(9)	-13.1(10)
C5	60.7(15)	48.0(13)	44.1(11)	3.7(10)	9.9(10)	-14.3(12)
C6	55.3(14)	50.2(13)	47.4(12)	-5(1)	12.4(10)	-15.9(11)
C7	50.5(14)	44.9(12)	49.2(12)	-8.6(10)	-0.5(10)	-9.8(11)
C8	58.3(15)	48.6(14)	65.5(14)	-8.9(12)	4.5(12)	-11.0(12)
C9	69.4(16)	45.7(14)	54.9(14)	-16.1(11)	-10.0(11)	2.4(12)
C10	56.3(14)	41.8(12)	40.6(11)	2.9(9)	5.6(9)	-15.5(11)
C11	89(2)	55.0(14)	37.8(11)	-2.2(10)	-8.9(11)	-8.1(13)
C12	81(2)	55.4(15)	61.2(14)	-8.2(11)	-27.6(13)	-1.3(13)
C13	77(2)	58.0(17)	94.7(19)	-11.6(14)	12.6(15)	-9.3(14)
C14	90(2)	85(2)	109(2)	14.2(17)	10.6(18)	-34.2(19)
C15	93(2)	130(3)	94(2)	-21(2)	18.0(17)	-69(2)
C16	95(3)	126(3)	109(3)	-48(2)	35.4(19)	-42(2)
C17	66(2)	77(2)	114(2)	-36.5(17)	2.1(16)	-6.3(15)
C18	76.3(17)	50.6(14)	44.8(12)	-4.9(10)	10.6(11)	-6.2(12)
C19	103(2)	47.4(14)	50.0(13)	-7.7(11)	0.6(12)	-15.7(14)
C20	60.0(17)	60.4(15)	74.9(16)	-2.3(12)	-12.0(12)	-15.5(13)
C21	62.9(18)	96(2)	85.0(18)	-13.6(16)	18.4(14)	-31.0(17)
C22	100(2)	102(2)	68.8(16)	4.3(16)	6.3(15)	-53(2)
C23	112(3)	64.1(17)	66.1(16)	15.7(13)	-22.0(15)	-23.6(17)
C24	84.7(19)	42.8(13)	65.6(14)	0.5(11)	-12.8(13)	-8.2(13)
N1	97.0(19)	83.0(17)	84.8(15)	-28.5(13)	29.6(14)	-15.7(14)

N2	128(2)	61.7(15)	56.0(12)	-6.0(11)	-16.0(13)	14.3(14)
N3	76.8(14)	41.1(10)	41.3(9)	5.5(8)	-1.9(9)	-17.3(10)
N4	53.6(12)	53.5(12)	60.3(11)	-13.3(9)	-6.4(9)	-7.2(9)
N5	64.0(13)	44.3(10)	42.1(9)	-1.0(8)	-0.9(8)	-8.2(9)
N6	56.6(12)	38.7(10)	47.6(9)	-1.3(8)	-2.8(8)	-7.4(9)

Table S1*d*. Bond Lengths for AEPiDQ.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
C1	C2	1.405(3)	C12	N4	1.453(3)
C1	C6	1.399(3)	C13	C14	1.497(3)
C1	C7	1.429(3)	C13	N4	1.447(3)
C2	C3	1.363(3)	C14	C15	1.510(4)
C3	C4	1.399(3)	C15	C16	1.504(4)
C4	C5	1.394(3)	C16	C17	1.509(4)
C4	C10	1.455(3)	C17	N4	1.456(3)
C5	C6	1.366(3)	C18	C19	1.500(3)
C7	C8	1.409(3)	C18	N5	1.454(3)
C7	C9	1.397(3)	C19	N6	1.468(2)
C8	N1	1.146(3)	C20	C21	1.511(3)

Table S1*e*. Bond Angles for AEPiDQ.

Atom	Atom	Atom	Angle/˚	Atom	Atom	Atom	Angle/˚
C2	C1	C7	121.54(18)	N4	C13	C14	111.5(2)
C6	C1	C2	116.28(19)	C13	C14	C15	111.5(3)
C6	C1	C7	122.19(17)	C16	C15	C14	109.9(2)
C3	C2	C1	122.13(18)	C15	C16	C17	111.5(2)
C2	C3	C4	121.06(18)	N4	C17	C16	110.4(2)
C3	C4	C10	121.60(17)	N5	C18	C19	114.91(19)
C5	C4	C3	117.21(19)	N6	C19	C18	113.25(17)
C5	C4	C10	121.01(17)	N6	C20	C21	111.23(19)

C6	C5	C4	121.59(19)	C22	C21	C20	110.6(2)
C5	C6	C1	121.71(18)	C21	C22	C23	110.7(2)
C8	C7	C1	121.83(18)	C22	C23	C24	111.8(2)
C9	C7	C1	120.59(18)	N6	C24	C23	110.84(18)
C9	C7	C8	117.6(2)	C10	N3	C11	123.69(17)
N1	C8	C7	178.6(3)	C12	N4	C17	111.2(2)
N2	C9	C7	177.3(3)	C13	N4	C12	113.35(18)
N3	C10	C4	122.04(18)	C13	N4	C17	110.1(2)
N5	C10	C4	118.21(18)	C10	N5	C18	125.53(18)
N5	C10	N3	119.60(19)	C20	N6	C19	111.45(18)
N3	C11	C12	110.41(17)	C20	N6	C24	110.12(18)
N4	C12	C11	112.23(18)	C24	N6	C19	110.93(16)

Table S1*f*. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for AEPiDQ

Atom	x	y	z	U(eq)
H2	504	10577	811	54
H3	2183	8465	859	54
H5	921	7770	3718	62
H6	-774	9879	3666	61
H11A	4115	6025	4494	75
H11B	3646	7397	3817	75
H12A	6367	6717	3997	82
H12B	6464	5328	3594	82
H13A	6553	8563	2817	94
H13B	4742	8704	2727	94
H14A	5680	9845	1349	114
H14B	5012	8704	1016	114
H15A	7386	8537	173	117
H15B	8248	8551	1162	117
H16A	8873	6345	787	127
H16B	7083	6430	662	127
H17A	7899	5277	2199	104

H17B	8464	6495	2495	104
H18A	4900	4471	1681	72
H18B	4209	4560	622	72
H19A	2400	3410	1297	81
H19B	4154	2535	1436	81
H20A	818	3103	2673	78
H20B	1003	4565	2698	78
H21A	1629	4114	4382	96
H21B	-18	3854	4234	96
H22A	1096	1567	4316	104
H22B	1645	2088	5251	104
H23A	3769	588	4440	98
H23B	4088	1989	4531	98
H24A	4786	1407	2924	79
H24B	3119	1187	2781	79
H3A	3495	4982	3258	64
H5A	2143	6206	1052	62

Table S2 Crystallographic data for Compound AEPyDQ

Table S2*a*. Crystal data and structure refinement for AEPyDQ.

Identification code	AEPyDQ
Empirical formula	$C_{24}H_{22}N_6$
Formula weight	394.47
Temperature/K	293(2)
Crystal system	orthorhombic
Space group	$P2_12_12_1$
a/Å	8.2134(11)
b/Å	12.2105(11)
c/Å	21.052(3)
α/°	90
β/°	90
γ/°	90
Volume/Å^3	2111.3(4)
Z	4
ρ_{calc}mg/mm^3	1.241
m/mm^{-1}	0.077
F(000)	832.0
Crystal size/mm^3	0.32 × 0.24 × 0.12
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection	5.324 to 52.736°
Index ranges	$-4 \le h \le 10$, $-9 \le k \le 15$, $-26 \le l \le 15$
Reflections collected	5955
Independent reflections	4005 [R_{int} = 0.0466, R_{sigma} = 0.1354]
Data/restraints/parameters	4005/0/271
Goodness-of-fit on F^2	0.951
Final R indexes [I>=2σ (I)]	R_1 = 0.0710, wR_2 = 0.0907
Final R indexes [all data]	R_1 = 0.1534, wR_2 = 0.1228
Largest diff. peak/hole / e Å^{-3}	0.17/-0.18
Flack parameter	-2.7(10)

Table S2*b*. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for AEPyDQ. U_{eq} is defined as one third of the trace of the orthogonalized U_{IJ} tensor.

Atom	x	y	z	U(eq)
C1	5301(7)	4747(3)	6105(2)	33.6(12)
C2	4614(7)	4895(4)	5501(2)	39.2(13)
C3	5552(7)	5165(4)	4981(2)	39.7(14)
C4	7213(7)	5313(4)	5044(2)	33.7(13)
C5	7932(7)	5145(3)	5636(2)	37.6(13)
C6	6984(7)	4861(4)	6151(2)	40.7(14)
C7	4280(7)	4473(4)	6641(3)	39.3(14)
C8	4965(7)	4215(4)	7232(3)	42.7(15)
C9	2613(9)	4381(5)	6582(3)	50.9(17)
C10	8197(7)	5706(4)	4508(2)	34.2(12)
C11	10455(7)	6969(4)	4195(2)	41.0(14)
C12	11938(7)	6258(4)	4220(3)	52.8(16)
C13	13351(7)	6692(4)	3842(3)	41.1(13)
C14	13696(8)	6335(5)	3231(3)	58.8(18)
C15	15030(10)	6747(6)	2923(3)	73(2)
C16	16006(8)	7483(5)	3219(3)	65.1(19)
C17	15592(8)	7807(4)	3820(3)	53.5(16)
C18	7317(7)	4234(4)	3780(2)	50.3(15)
C19	8465(8)	3534(4)	3386(3)	56.1(18)
C20	10068(9)	3340(4)	3703(3)	53.8(18)
C21	11451(11)	3258(5)	3349(4)	87(3)
C22	12902(14)	3038(8)	3652(6)	126(5)
C23	12936(15)	2928(6)	4289(6)	119(4)
C24	11484(13)	3046(6)	4607(4)	96(3)
N1	5540(7)	3992(4)	7715(2)	62.1(15)
N2	1223(7)	4308(5)	6515(3)	82.2(19)
N3	9188(5)	6550(3)	4619.6(18)	38.1(11)
N4	14309(6)	7425(3)	4133(2)	44.1(12)

N5	8069(6)	5288(3)	3933.2(17)	39.8(11)
N6	10051(8)	3245(4)	4330(3)	75.7(18)

Table S2*c*. Anisotropic Displacement Parameters ($Å^2×10^3$) for AEPyDQ. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	U11	U22	U33	U23	U13	U12
C1	36(3)	35(3)	30(3)	4(2)	0(3)	0(3)
C2	32(3)	46(3)	40(3)	6(3)	-4(3)	-1(3)
C3	42(4)	42(3)	35(3)	7(3)	-5(3)	0(3)
C4	34(3)	38(3)	30(3)	-2(2)	1(3)	0(3)
C5	31(3)	44(3)	38(3)	4(2)	-3(3)	-5(3)
C6	45(4)	46(3)	31(3)	8(3)	-8(3)	0(3)
C7	38(4)	47(3)	32(3)	5(2)	0(3)	0(3)
C8	45(4)	50(3)	33(3)	-2(3)	7(3)	-6(3)
C9	49(4)	63(4)	40(4)	21(3)	7(4)	6(3)
C10	34(3)	36(3)	33(3)	-2(2)	2(3)	7(3)
C11	46(4)	41(3)	35(3)	-3(2)	5(3)	-4(3)
C12	47(4)	53(3)	59(4)	9(3)	6(4)	3(3)
C13	34(3)	48(3)	42(3)	2(3)	1(3)	5(3)
C14	50(5)	82(4)	44(4)	-16(3)	-3(4)	-14(4)
C15	73(6)	107(5)	39(4)	-19(4)	10(4)	-2(5)
C16	51(5)	89(5)	55(4)	1(4)	13(4)	-8(4)
C17	51(4)	49(3)	61(4)	-6(3)	-1(4)	-11(3)
C18	52(4)	50(3)	49(3)	-10(3)	0(4)	-13(3)
C19	69(5)	52(3)	48(4)	-17(3)	4(4)	-5(3)
C20	55(5)	41(3)	66(5)	-16(3)	4(4)	-7(3)
C21	70(6)	85(5)	105(6)	-52(5)	12(6)	-11(5)
C22	67(7)	118(7)	193(12)	-90(9)	18(10)	-8(6)
C23	73(7)	63(5)	220(13)	-21(7)	-36(11)	2(5)
C24	99(7)	77(5)	113(7)	28(5)	-37(7)	-17(6)
N1	64(4)	88(3)	34(3)	7(3)	-4(3)	-9(3)
N2	46(4)	129(5)	72(4)	46(4)	-3(4)	1(4)

N3	43(3)	43(2)	29(2)	-6.2(19)	5(2)	-6(2)
N4	46(3)	46(2)	41(3)	0(2)	7(3)	-3(3)
N5	52(3)	37(2)	30(2)	-5.6(19)	2(3)	-7(2)
N6	79(5)	75(4)	72(4)	16(3)	-11(4)	-9(4)

Table S2*d*. Bond Lengths for AEPyDQ.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
C1	C2	1.403(6)	C12	C13	1.502(7)
C1	C6	1.393(7)	C13	C14	1.389(7)
C1	C7	1.445(7)	C13	N4	1.339(6)
C2	C3	1.378(6)	C14	C15	1.369(8)
C3	C4	1.383(7)	C15	C16	1.356(8)
C4	C5	1.395(6)	C16	C17	1.367(7)
C4	C10	1.468(6)	C17	N4	1.328(7)
C5	C6	1.379(6)	C18	C19	1.519(7)
C7	C8	1.401(7)	C18	N5	1.463(5)
C7	C9	1.379(9)	C19	C20	1.495(8)
C8	N1	1.154(6)	C20	C21	1.362(9)
C9	N2	1.154(8)	C20	N6	1.324(8)
C10	N3	1.334(6)	C21	C22	1.378(12)
C10	N5	1.317(5)	C22	C23	1.349(12)
C11	C12	1.497(7)	C23	C24	1.374(12)
C11	N3	1.464(6)	C24	N6	1.336(10)

Table S2*e*. Bond Angles for AEPyDQ

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C2	C1	C7	120.3(5)	C14	C13	C12	122.5(5)
C6	C1	C2	116.7(5)	N4	C13	C12	116.6(5)
C6	C1	C7	123.0(5)	N4	C13	C14	120.9(5)
C3	C2	C1	121.7(5)	C15	C14	C13	119.2(6)
C2	C3	C4	120.5(5)	C16	C15	C14	119.9(6)
C3	C4	C5	118.9(5)	C15	C16	C17	118.0(6)
C3	C4	C10	120.8(5)	N4	C17	C16	123.7(6)

C5	C4	C10	120.2(5)	N5	C18	C19	110.7(5)
C6	C5	C4	120.1(5)	C20	C19	C18	113.1(5)
C5	C6	C1	122.0(5)	C21	C20	C19	120.1(7)
C8	C7	C1	120.8(5)	N6	C20	C19	116.6(7)
C9	C7	C1	121.6(6)	N6	C20	C21	123.2(8)
C9	C7	C8	117.4(6)	C20	C21	C22	118.8(9)
N1	C8	C7	179.1(6)	C23	C22	C21	119.9(13)
N2	C9	C7	178.1(8)	C22	C23	C24	117.1(13)
N3	C10	C4	116.9(4)	N6	C24	C23	124.8(9)
N5	C10	C4	122.4(5)	C10	N3	C11	126.6(4)
N5	C10	N3	120.7(5)	C17	N4	C13	118.3(5)
N3	C11	C12	110.8(4)	C10	N5	C18	125.2(4)
C11	C12	C13	113.9(4)	C20	N6	C24	116.2(7)

Table S2*f*. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for AEPyDQ

Atom	x	y	z	U(eq)
H2	3496	4808	5450	47
H3	5064	5248	4585	48
H5	9051	5224	5684	45
H6	7484	4743	6542	49
H11A	10741	7709	4320	49
H11B	10042	6993	3764	49
H12A	11658	5537	4061	63
H12B	12272	6178	4659	63
H14	13029	5823	3033	71
H15	15268	6521	2511	88
H16	16928	7761	3019	78
H17	16247	8323	4020	64
H18A	6318	4357	3545	60
H18B	7045	3852	4170	60

H19A	7950	2833	3303	67
H19B	8650	3890	2981	67
H21	11416	3349	2910	104
H22	13855	2966	3417	151
H23	13899	2779	4504	142
H24	11501	2983	5047	116
H3A	9071	6881	4977	46
H5A	8458	5665	3623	48

Table S3 Crystallographic data for Compound AEPrDQ

Table S3*a*. Crystal data and structure refinement for AEPrDQ.

Identification code	AEPrDQ
Empirical formula	$C_{22}H_{30}N_6$
Formula weight	378.52
Temperature/K	293.15
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	13.1430(12)
b/Å	10.3517(9)
c/Å	16.1607(15)
α/°	90
β/°	101.6510(10)
γ/°	90
Volume/Å^3	2153.4(3)
Z	4
ρ_{calc}mg/mm^3	1.168
m/mm^{-1}	0.072
F(000)	816.0
Crystal size/mm^3	0.32 × 0.18 × 0.12
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection	3.164 to 51.892°
Index ranges	$-16 \le h \le 16$, $-12 \le k \le 12$, $-19 \le l \le 19$
Reflections collected	21689
Independent reflections	4206 [R_{int} = 0.0306, R_{sigma} = 0.0230]
Data/restraints/parameters	4206/1/253
Goodness-of-fit on F^2	1.031
Final R indexes [I>=2σ (I)]	R_1 = 0.0511, wR_2 = 0.1391
Final R indexes [all data]	R_1 = 0.0752, wR_2 = 0.1573
Largest diff. peak/hole / e Å^{-3}	0.16/-0.18

Table S3*b*. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters (Å^2×10^3) for AEPrDQ. U_{eq} is defined as one third of the trace of the orthogonalized U_{IJ} tensor.

Atom	x	y	z	U(eq)
C1	10978.0(12)	855.5(15)	7429.9(10)	51.7(4)
C2	10381.3(13)	1987.4(15)	7266.6(10)	56.5(4)
C3	9611.4(13)	2107.1(15)	6565.8(10)	57.3(4)

C4	9375.4(12)	1108.5(15)	5978.8(10)	53.0(4)
C5	9964.1(13)	-19.4(15)	6134.4(10)	55.7(4)
C6	10741.9(13)	-139.2(15)	6831.8(11)	55.3(4)
C7	11778.2(13)	727.8(15)	8176.7(11)	58.1(4)
C8	11938.6(15)	1718.5(18)	8777.7(12)	69.2(5)
C9	12402.5(14)	-379.1(19)	8327.2(12)	67.1(5)
C10	8490.6(13)	1205.7(16)	5268.6(10)	54.6(4)
C11	8948.3(15)	3384.3(18)	4834.6(11)	67.8(5)
C12	8936.9(14)	3843.7(17)	3953.6(11)	64.4(5)
C13	7794.6(19)	5709(2)	3866.7(14)	87.0(6)
C14	7033(2)	6350(2)	3165.1(18)	109.8(8)
C15	7014(2)	5539(3)	2392.4(16)	109.8(8)
C16	7897.6(17)	4620(2)	2645.7(12)	77.9(6)
C17	7098.2(16)	-19(2)	4318.2(14)	82.8(6)
C18	6078.9(16)	581(3)	4385.6(16)	96.7(7)
C19	5722(2)	2275(3)	3323.4(17)	109.7(9)
C20	5037(3)	3432(4)	3279(3)	164.6(16)
C21	5123(3)	3862(4)	4145(4)	191(2)
C22	5330(3)	2659(4)	4647(2)	150.1(13)
N1	12029.9(18)	2558.3(18)	9248.4(12)	101.5(7)
N2	12884.7(15)	-1310.3(19)	8431.4(13)	101.0(6)
N3	8253.3(11)	2285.1(13)	4839.8(9)	59.1(4)
N4	7963.3(11)	4427.9(13)	3550.9(9)	60.0(4)
N5	7911.5(11)	165.9(14)	5063.4(10)	66.3(4)
N6	6048.4(12)	1948.2(19)	4214.7(11)	83.6(5)

Table S3*c*. Anisotropic Displacement Parameters ($Å^2 \times 10^3$) for AEPrDQ. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	U11	U22	U33	U23	U13	U12
C1	54.9(9)	48.7(9)	52.5(9)	2.8(7)	13.4(7)	-4.0(7)
C2	69.6(10)	48.1(9)	51.7(9)	-2.4(7)	11.9(8)	2.3(8)
C3	65.9(10)	50.2(9)	56.1(10)	3.7(7)	13.2(8)	8.1(8)
C4	57.9(9)	52.6(9)	48.9(9)	3.5(7)	11.7(7)	-0.3(7)
C5	63.4(10)	50.8(9)	53.2(9)	-4.0(7)	12.7(8)	-1.2(7)
C6	58.7(10)	46.8(9)	59.6(10)	0.1(7)	10.3(8)	4.1(7)
C7	62.9(10)	49.0(9)	59.3(10)	1.2(7)	5.1(8)	-1.7(8)
C8	84.8(13)	57.1(11)	58.3(11)	6.3(9)	-3.1(9)	1.1(9)
C9	62.4(11)	60.3(11)	71.8(12)	-0.4(9)	-2.7(9)	-5.3(9)
C10	55.8(9)	54.3(10)	54.5(9)	2.5(7)	13.3(7)	-0.6(7)
C11	68.9(11)	61.4(11)	67.4(11)	9.0(9)	0.3(9)	-9.4(9)
C12	66.4(11)	56.4(10)	70.6(11)	5.5(8)	14.7(9)	-4.1(8)
C13	100.6(16)	69.3(13)	89.9(15)	-9.8(11)	16.2(12)	9.4(11)
C14	115(2)	84.4(16)	129(2)	22.8(15)	19.9(16)	31.8(14)
C15	114(2)	128(2)	85.6(17)	36.4(16)	15.4(14)	24.7(17)
C16	93.0(14)	79.4(13)	61.4(11)	8.4(10)	16(1)	2.1(11)
C17	79.5(14)	67.6(12)	90.4(15)	1.3(10)	-8.5(11)	-17(1)
C18	65.3(13)	110.4(19)	108.3(18)	26.6(14)	3.1(12)	-21.3(12)
C19	94.9(17)	116(2)	102.8(19)	18.5(15)	-17.2(14)	-19.2(15)
C20	138(3)	115(3)	209(4)	42(3)	-40(3)	-2(2)
C21	110(3)	143(3)	288(6)	-57(4)	-35(3)	33(2)
C22	93(2)	201(4)	151(3)	-43(3)	11.9(19)	23(2)
N1	146.9(18)	70.8(11)	71.3(11)	-11.7(9)	-14.8(11)	13.0(11)
N2	87.5(13)	76.0(12)	123.5(16)	-5.0(11)	-16.4(11)	18.4(10)
N3	54.5(8)	57.4(8)	61.2(8)	8.8(6)	1.5(6)	-4.4(6)
N4	69.1(9)	52.3(8)	57.3(8)	2.6(6)	9.7(7)	-0.4(7)
N5	68.2(9)	57.7(9)	67.9(9)	7.1(7)	1.9(7)	-8.4(7)
N6	59.7(10)	95.5(13)	90.2(12)	-0.4(10)	2.1(9)	-1.2(9)

Table S3*d*. Bond Lengths for AEPrDQ.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
C1	C2	1.405(2)	C11	N3	1.460(2)
C1	C6	1.403(2)	C12	N4	1.446(2)
C1	C7	1.438(2)	C13	C14	1.507(3)
C2	C3	1.363(2)	C13	N4	1.454(2)
C3	C4	1.395(2)	C14	C15	1.501(4)
C4	C5	1.395(2)	C15	C16	1.493(3)
C4	C10	1.463(2)	C16	N4	1.461(2)
C5	C6	1.366(2)	C17	C18	1.500(3)
C7	C8	1.399(3)	C17	N5	1.452(2)
C7	C9	1.402(3)	C18	N6	1.441(3)
C8	N1	1.145(2)	C19	C20	1.490(4)
C9	N2	1.147(2)	C19	N6	1.457(3)
C10	N3	1.319(2)	C20	C21	1.452(6)
C10	N5	1.321(2)	C21	C22	1.481(6)
C11	C12	1.498(2)	C22	N6	1.480(4)

Table S3*e*. Bond Angles for AEPrDQ

Atom	Atom	Atom	Angle/˚	Atom	Atom	Atom	Angle/˚
C2	C1	C7	121.35(14)	N4	C12	C11	113.49(15)
C6	C1	C2	116.43(14)	N4	C13	C14	105.23(18)
C6	C1	C7	122.22(14)	C15	C14	C13	106.06(19)
C3	C2	C1	121.62(15)	C16	C15	C14	104.3(2)
C2	C3	C4	121.56(15)	N4	C16	C15	104.27(18)
C3	C4	C5	117.29(14)	N5	C17	C18	113.8(2)
C3	C4	C10	120.99(14)	N6	C18	C17	112.53(17)
C5	C4	C10	121.53(14)	N6	C19	C20	106.8(3)
C6	C5	C4	121.34(15)	C21	C20	C19	105.7(3)
C5	C6	C1	121.75(15)	C20	C21	C22	103.9(3)
C8	C7	C1	119.83(15)	N6	C22	C21	102.9(3)
C8	C7	C9	118.56(15)	C10	N3	C11	125.55(14)

C9	C7	C1	121.61(15)	C12	N4	C13	114.05(15)
N1	C8	C7	176.8(2)	C12	N4	C16	112.04(15)
N2	C9	C7	177.5(2)	C13	N4	C16	104.19(15)
N3	C10	C4	122.46(14)	C10	N5	C17	127.26(16)
N3	C10	N5	119.69(15)	C18	N6	C19	114.1(2)
N5	C10	C4	117.85(14)	C18	N6	C22	113.2(2)
N3	C11	C12	111.50(14)	C19	N6	C22	105.2(2)

Table S3*f*. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for AEPrDQ

Atom	x	y	z	U(eq)
H2	10515	2672	7646	68
H3	9235	2873	6477	69
H5	9824	-704	5756	67
H6	11125	-900	6913	66
H11A	9650	3136	5100	81
H11B	8737	4085	5161	81
H12A	9490	4469	3969	77
H12B	9078	3117	3615	77
H13A	8441	6190	3994	104
H13B	7511	5652	4375	104
H14A	6348	6384	3302	132
H14B	7254	7224	3074	132
H15A	6361	5079	2236	132
H15B	7112	6069	1920	132
H16A	7760	3810	2342	93
H16B	8538	4984	2537	93
H17A	6996	-938	4217	99
H17B	7326	348	3834	99
H18A	5526	160	3989	116
H18B	5955	438	4950	116
H19A	5345	1560	3015	132
H19B	6321	2467	3079	132

H20A	5265	4105	2940	198
H20B	4324	3209	3031	198
H21A	4483	4265	4224	229
H21B	5689	4473	4302	229
H22A	5652	2844	5229	180
H22B	4695	2175	4634	180
H3A	7637	2347	4538	71
H5A	8029	-473	5410	80

Figure S1 **Fig. 1** Absorption and the emission spectra of **(a)** AEPiDQ (------, magenta) **(b)** AEPyDQ (……., green) and **(c)** AEPrDQ (-.-.-., grey) respectively.

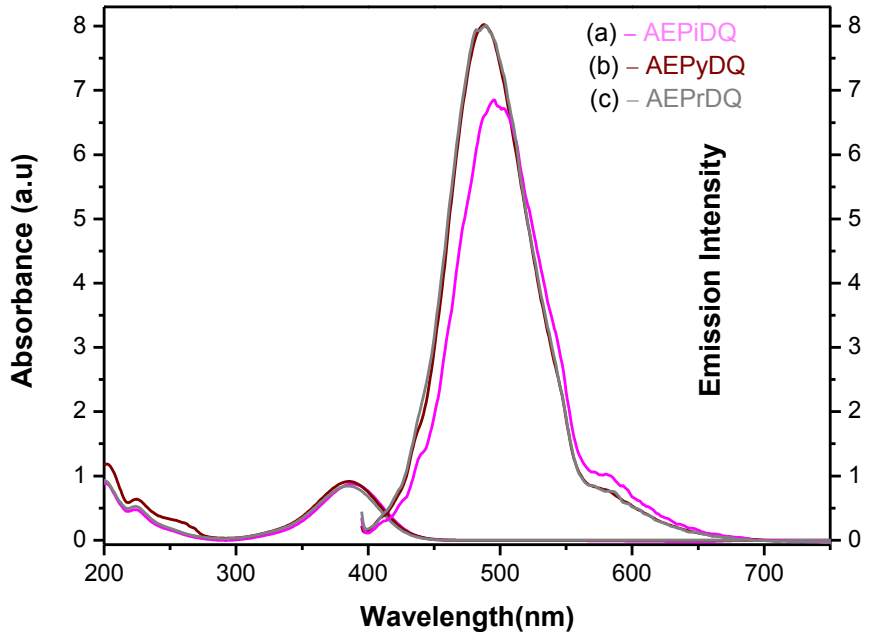


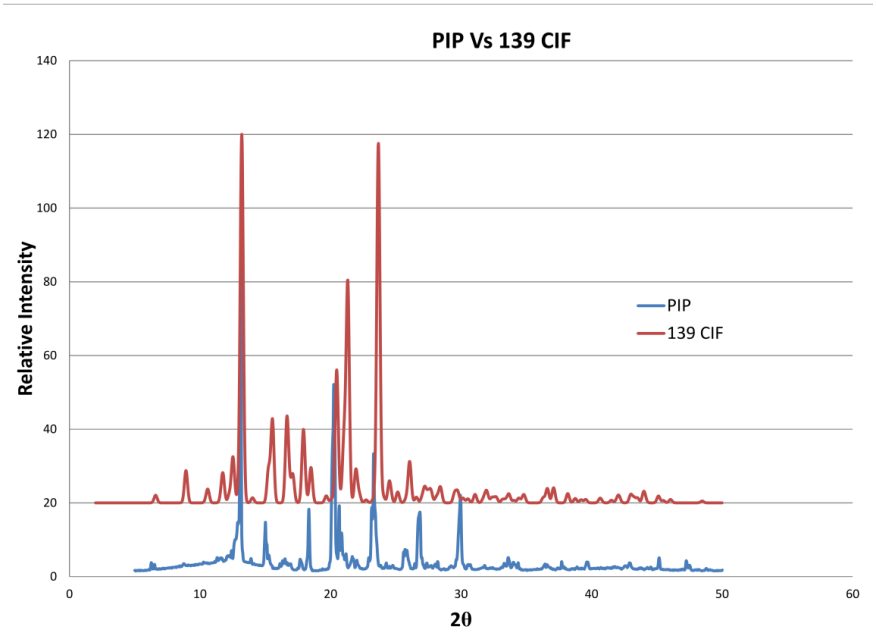
Table S4 Absorption and emission λ_{max} for AEPiDQ, AEPyDQ and AEPrDQ in acetonitrile.

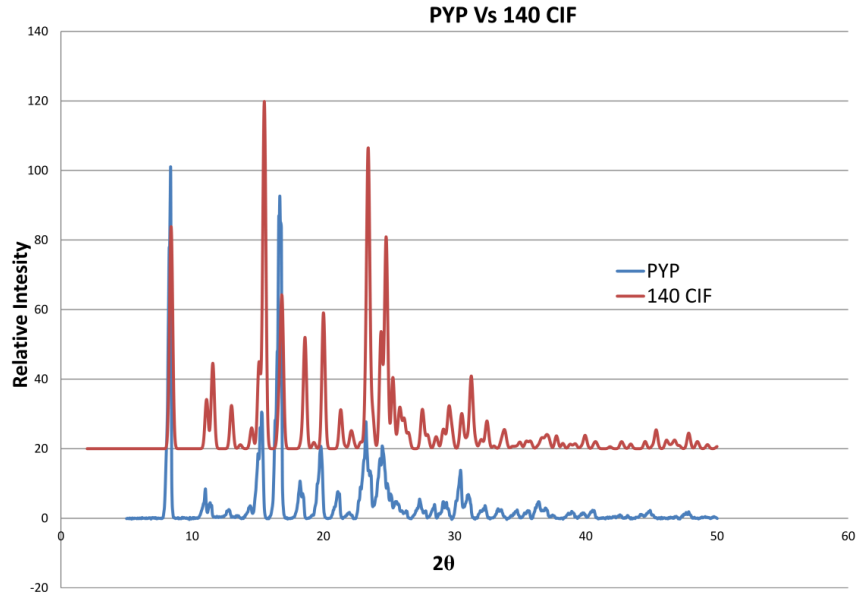
S. No.	Solutions	Absorption λ_{max} (nm)			Emission λ_{max} (nm)		
		AEPiDQ	AEPyDQ	AEPrDQ	AEPiDQ	AEPyDQ	AEPrDQ
1.	Acetonitrile	385	385	385	488	495	489

Table S5 Powder SHG data showing phase-matchable behavior in *Bis*- (2-(2-Amino ethyl)pyridino) dicyanoquinodimethane (AEPyDQ)

Size of the compound (μ)	NLO activity (Mv)
355	62
300	84
250	86
200	105
150	129
100	117
40	130

Figure S2 Comparison of powder X-ray data with the single X-ray crystal structure CIF files: PIP, PYP and PYR refers to AEPiDQ, AEPyDQ and AEPrDQ.




PYP Vs 140 CIF
Relative Intesity
2θ
PYP
140 CIF
140
120
100
80
60
40
20
0
-20
0
10
20
30
40
50
60

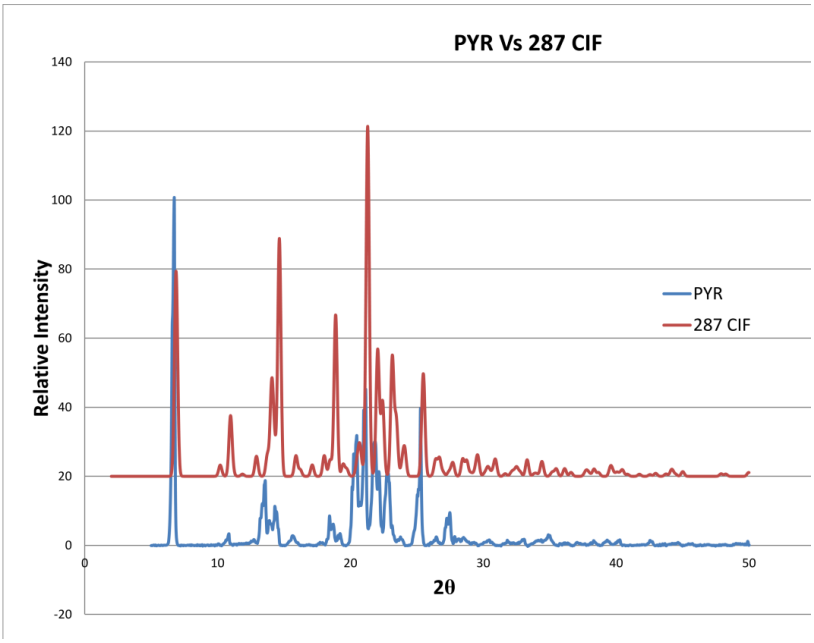
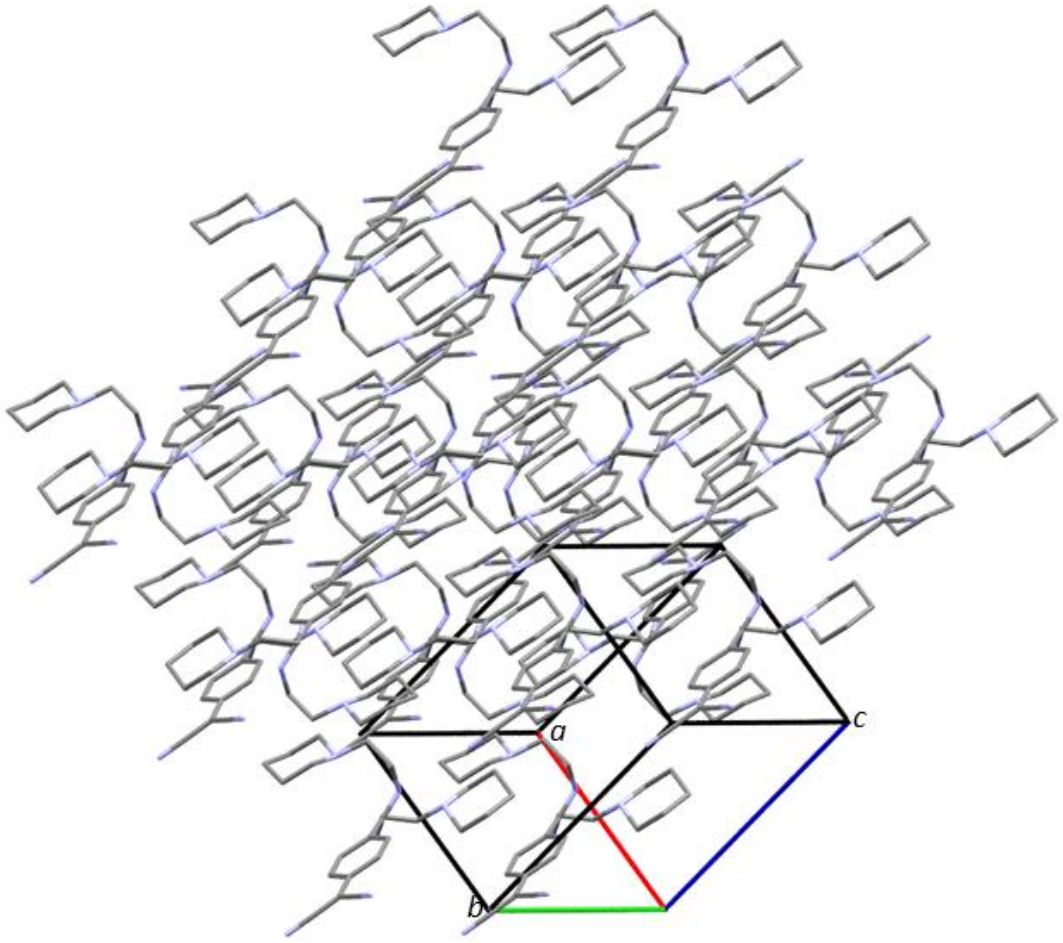
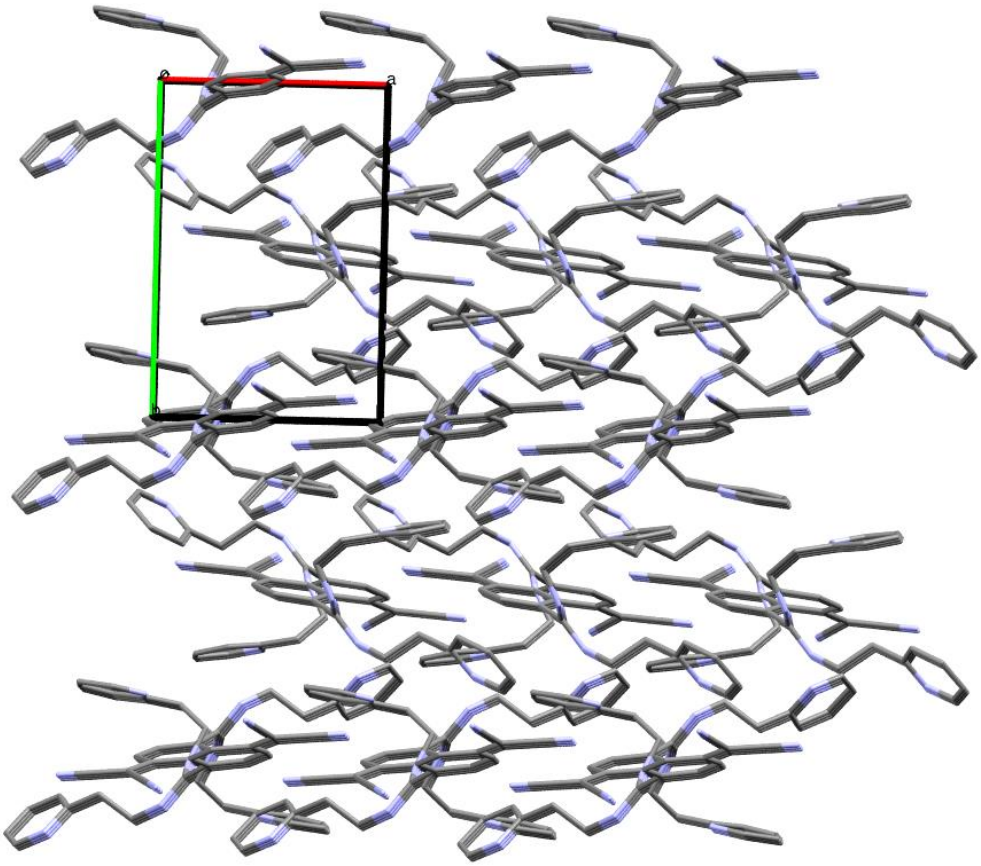

PYR Vs 287 CIF
Relative Intensity
2θ
PYR
287 CIF
140
120
100
80
60
40
20
0
-20
0
10
20
30
40
50

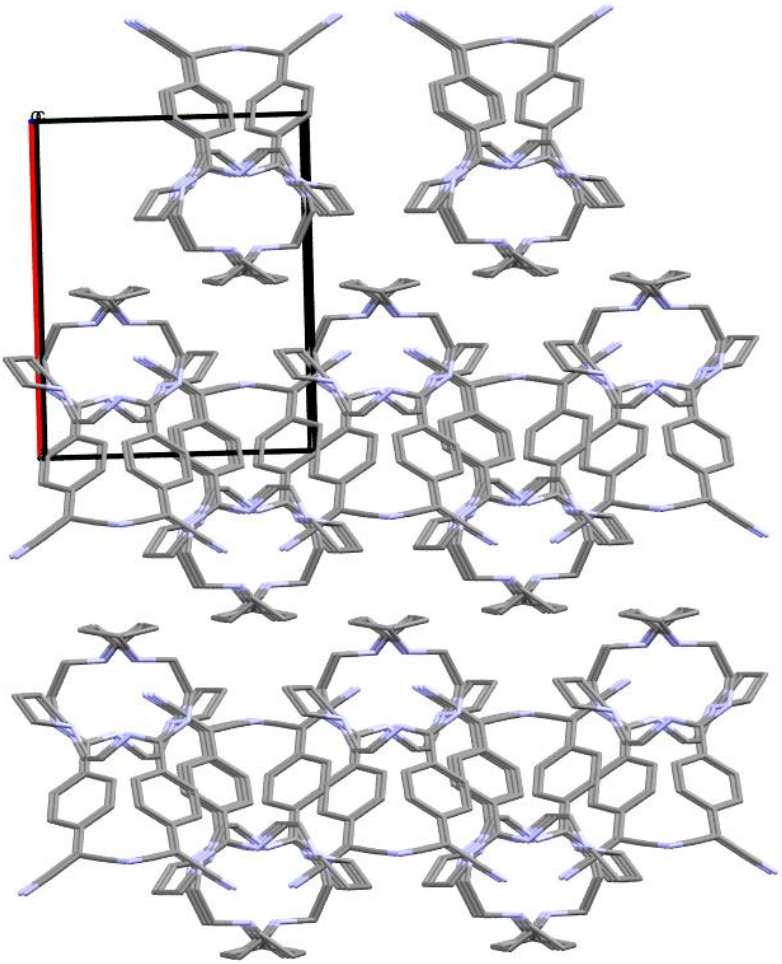
Figure S3 Supramolecular structures and molecular packing view, with arrangement of dipoles in a distinct fashion in (a) AEPiDQ (b) AEPyDQ and AEPrDQ. Hydrogen atoms are omitted for clarity.



(a)



(b)



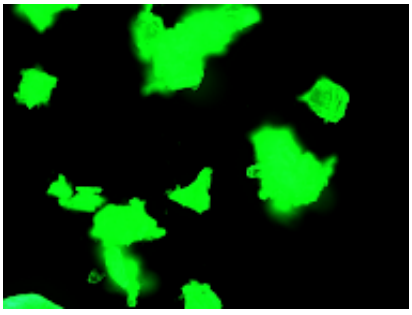
(c)

Figure S4 Yellow to pale yellow color observed under ambient light (a), (b), (c) and bright green emission noted under fluorescence microscope images acquired with a 10X objective lens (d), (e) and (f) respectively for AEPiDQ, AEPyDQ and AEPrDQ.

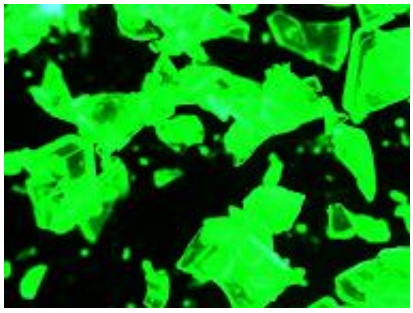
(a)

(b)

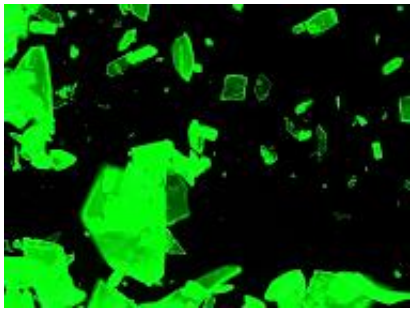
(c)



(d)



(e)



(f)