Modeling and Tuning the Electronic, Mechanical and Optical Properties of Recently Synthesized 2D Polyaramids: A First Principles Study

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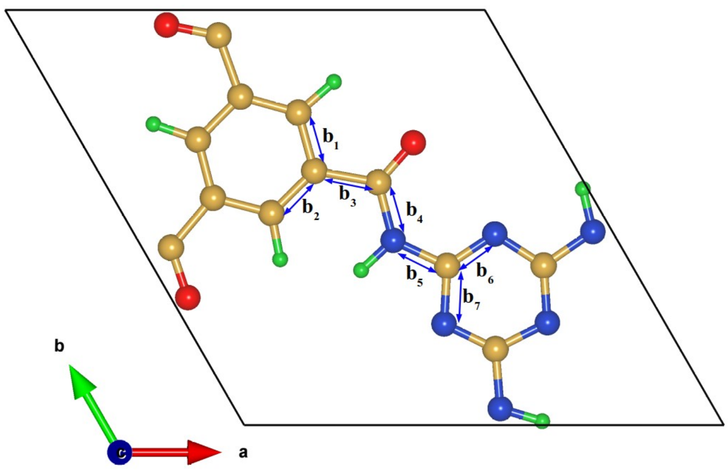

Fig. S1: Names of bonds in geometrically optimized 2DPA-1 structure are shown with double arrows. In the case of NN-2DPA and BN-2DPA structures, we replace the name of bonds with superscript NN and BN, respectively. For example, b_1^{NN}, b_1^{BN} represent names of bonds in place of b1 in NN and BN-2DPA structures.

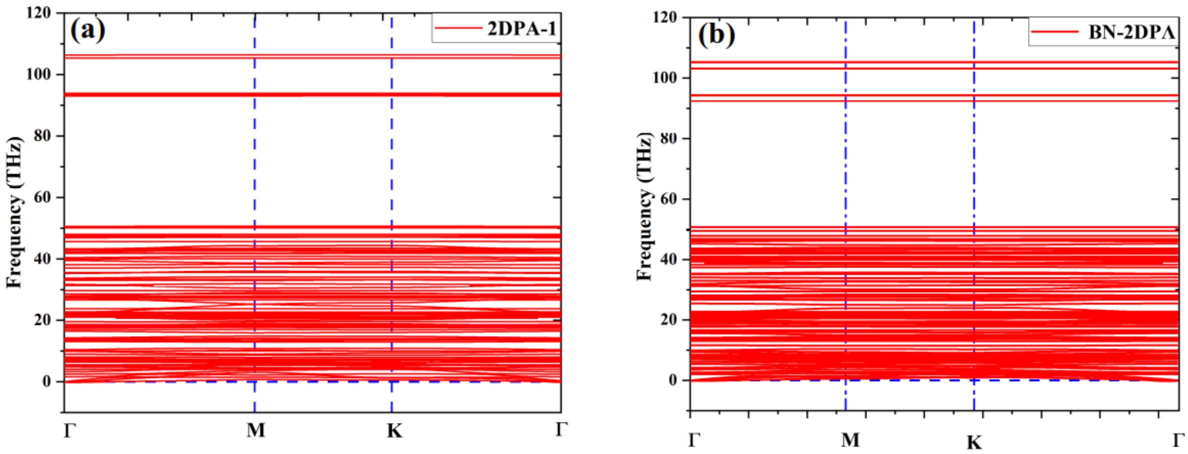

Fig. S2: Full Phonon spectra of (a) 2DPA-1, (b) BN-2DPA systems.

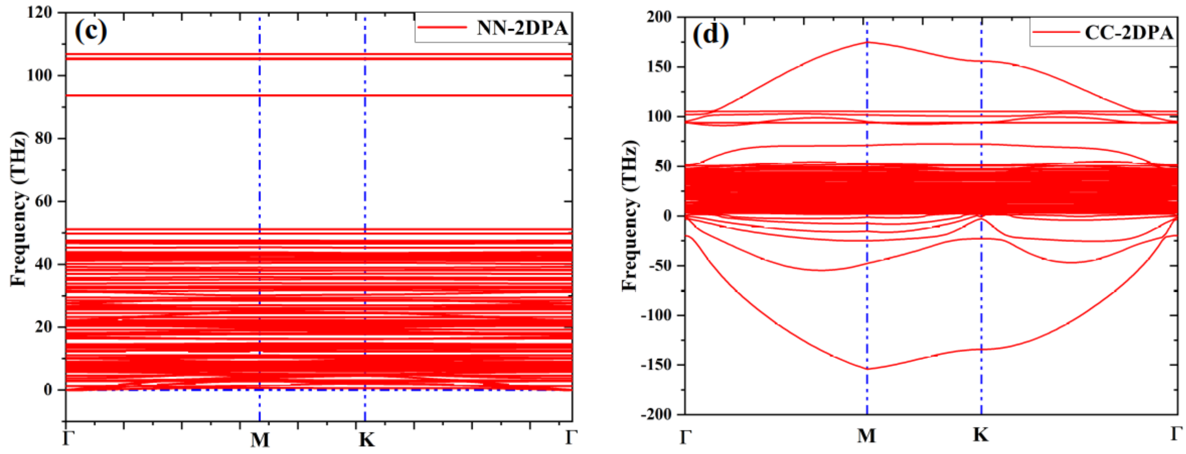

Fig. S3: Full Phonon spectra of (a) NN-2DPA, (b) CC-2DPA systems.

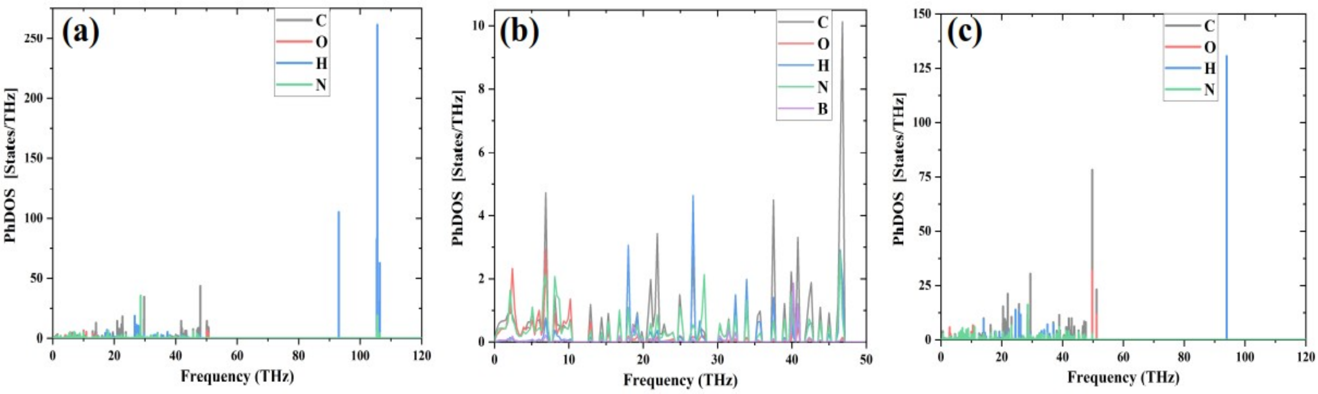

Fig. S4: Full Ph-DOS in the range of (0-20) THz of (a) 2DPA-1, (b) BN-2DPA, and (c) NN-2DPA systems.

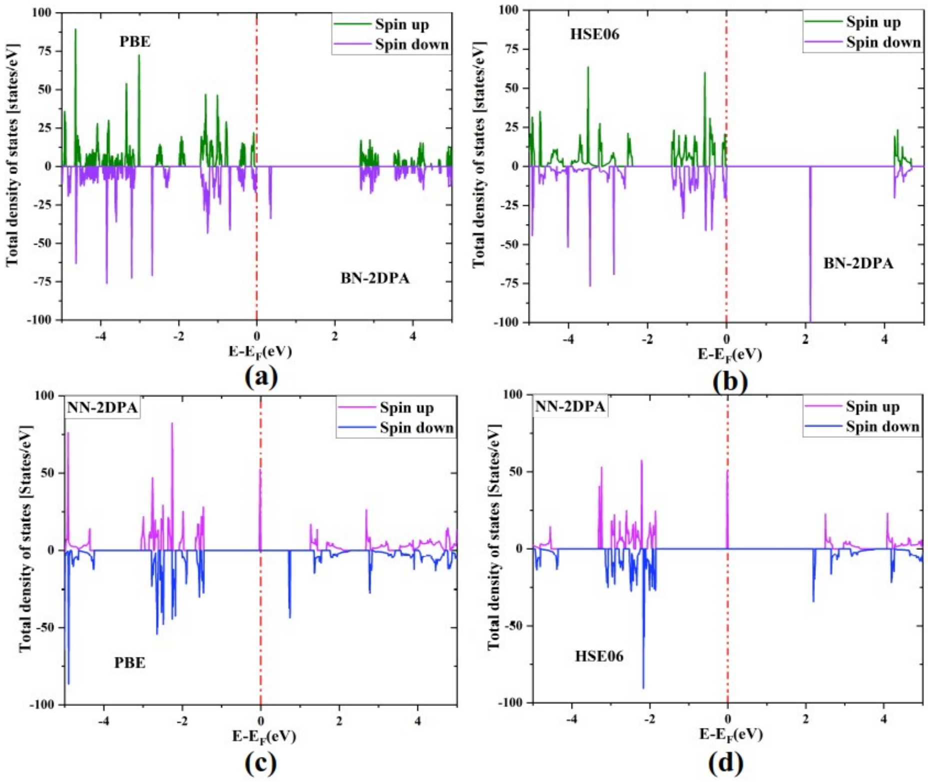

Fig. S5: Spin-polarized total density of states of (a) BN-2DPA using PBE, (b) BN-2DPA using HSE06, (c) NN-2DPA using PBE, (d) NN-2DPA using HSE06

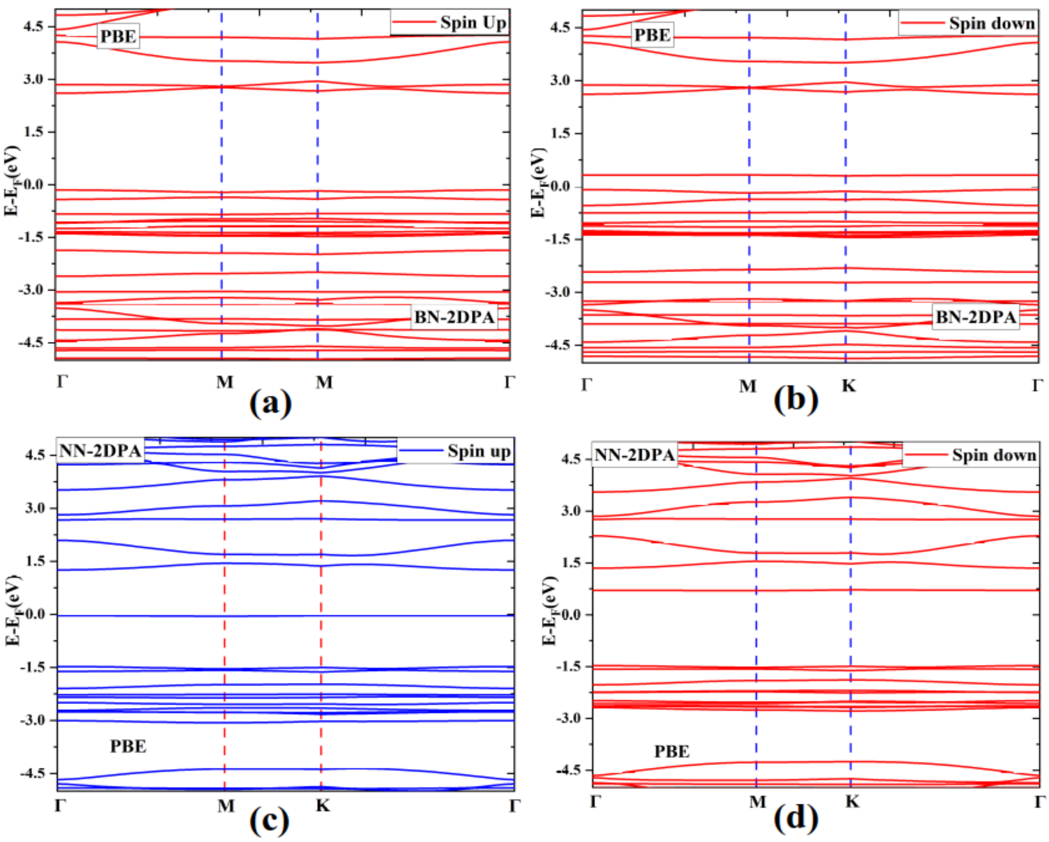

Fig. S6: Using PBE functional, spin-polarized band structure of (a) BN-2DPA for spin up channel, (b) BN-2DPA for spin down channel, (c) NN-2DPA for spin up channel, (d) NN-2DPA for spin down channel

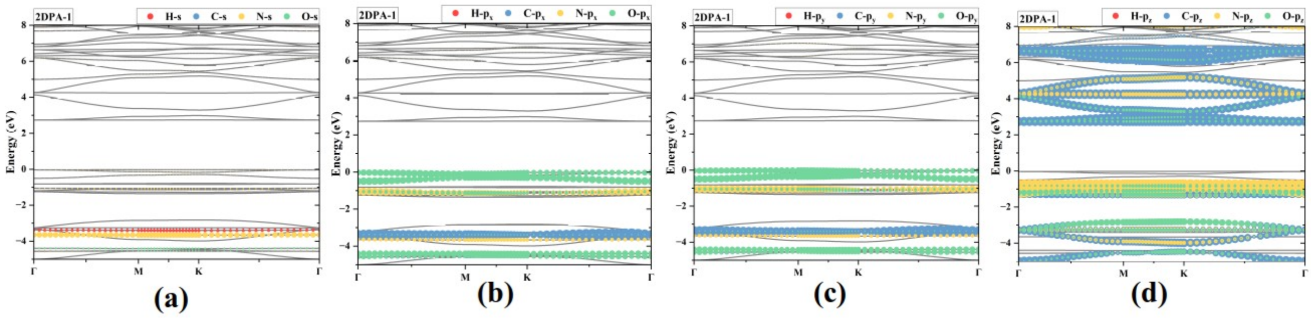

Fig. S7: Using PBE functional projected band structures of each different atom of 2DPA-1 with orbital (a) s, (b) px, (c) py, (d) pz.

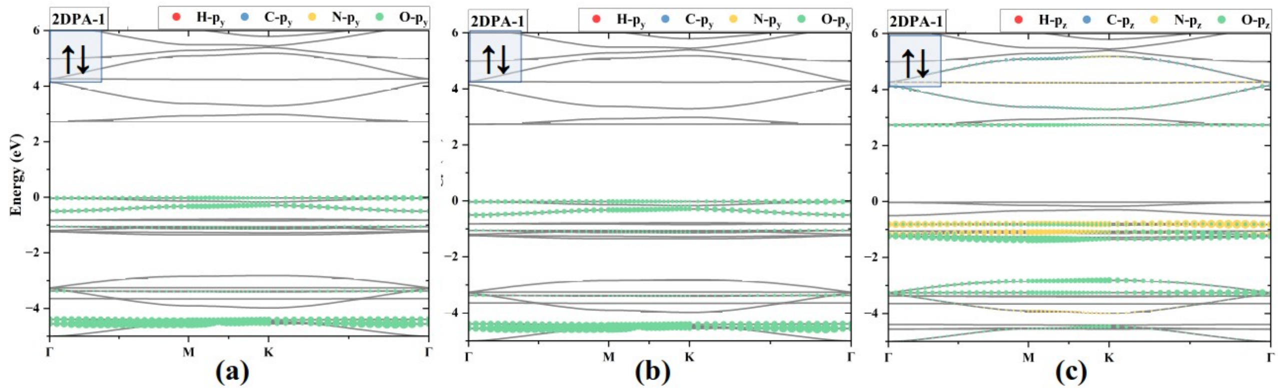

Fig. S8: Using PBE functional p-orbital projected band structures of 2DPA-1 for amide group and its nearest H and O atoms. The up/down arrows indicate spin-up/down.

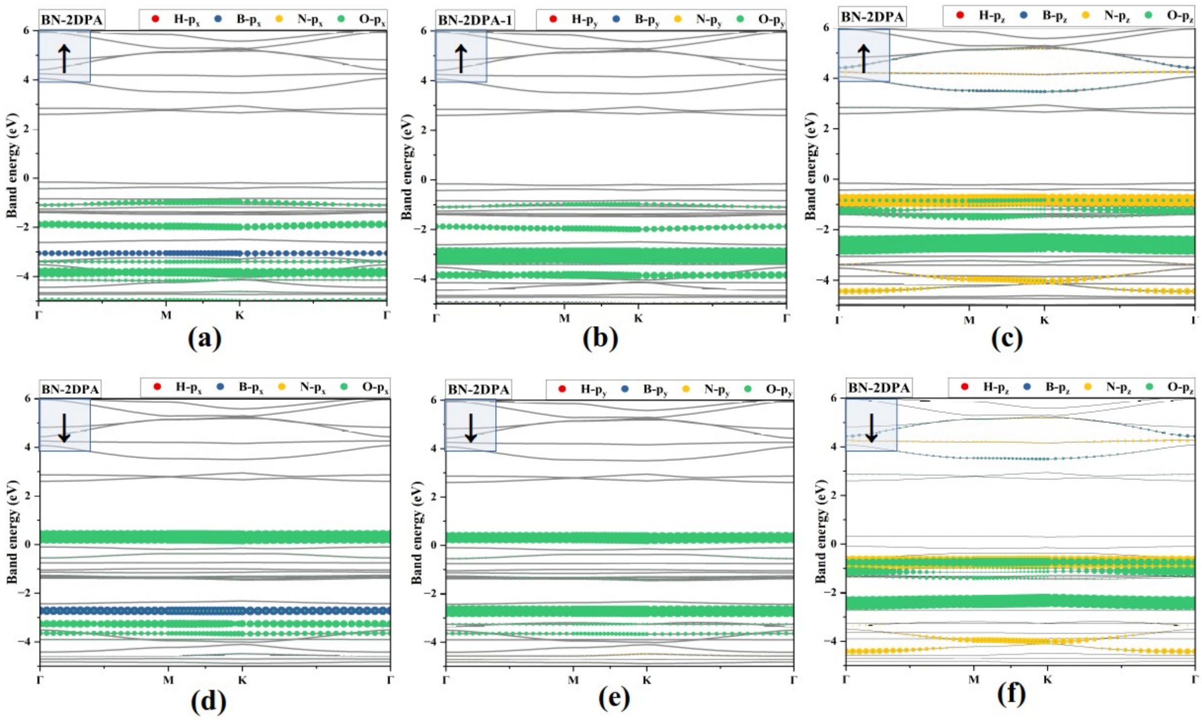

Fig. S9: Using PBE functional p-orbital projected band structures of BN-2DPA for BN group and its nearest H and O atoms with spin-up (a-c) and spin-down (d-f). The up/down arrows indicate spin-up/down.

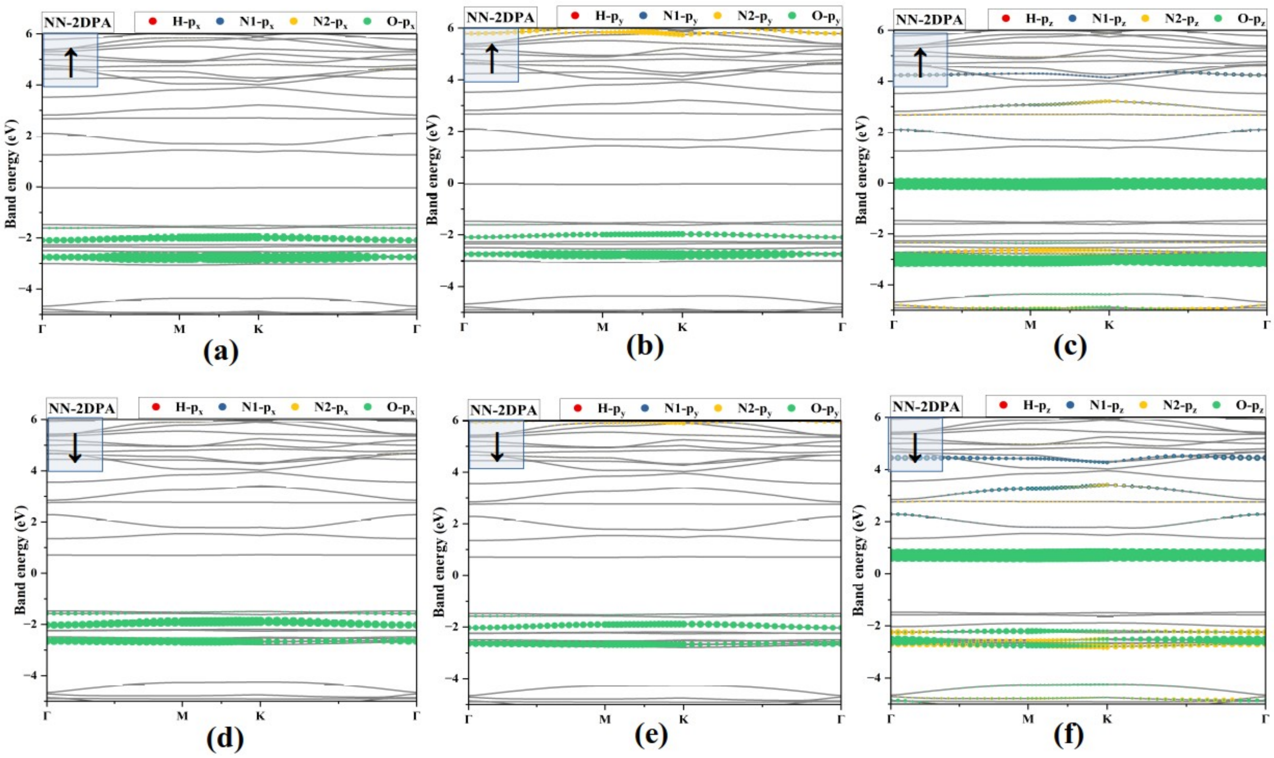

Fig. S10: Using PBE functional p-orbital projected band structures of NN-2DPA for NN group and its nearest H and O atoms with spin-up (a-c) and spin-down (d-f). The up/down arrows indicate spin-up/down.

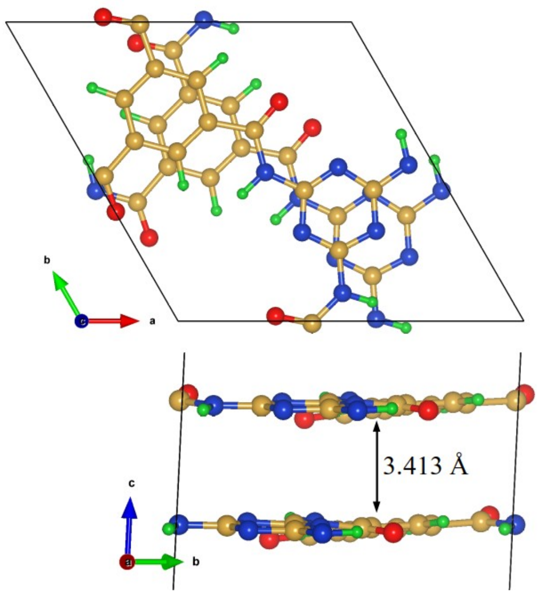

Fig. S11: Top and side vies of double layered 2DPA-1 optimized structure with PBE+Grimee-D3.

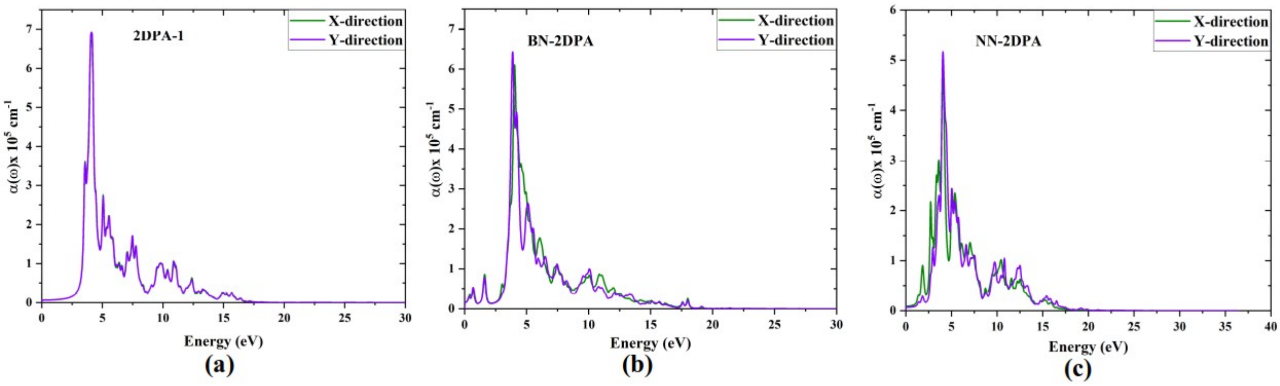

Fig. S12: Optical absorption spectra of (a) 2DPA-1, (b) BN-2DPA, and (c) NN-2DPA using the PBE functional.

Table S1: Geometrical optimized parameters of 2DPA-1, BN, and NN-2DPA systems. The names of the bonds are indicated in Fig. S1.

S.N.	System	Bond-length (Å)	Band gap (eV)	
			PBE	**HSE06**
1.	2DPA-1	b_1=1.40, b_2=1.40, b_3=1.51, b_4=1.39, b_5=1.39, b_6=1.33, b_7=1.35	2.76	4.42
2.	BN-2DPA	b_1^{BN}=1.40, b_2^{BN}=1.40, b_3^{BN}=1.59, b_4^{BN}=1.44, b_5^{BN}=1.35, b_6^{BN}=1.34, b_7^{BN}=1.34	0.32	2.13
3.	NN-2DPA	b_1^{NN}=1.41, b_2^{NN}=1.40, b_3^{NN}=1.42, b_4^{NN}=1.39, b_5^{NN}=1.38, b_6^{NN}=1.33, b_7^{NN}=1.36	0.74	2.19

Table S2: Cohesive energy comparison with different 2D materials composed of carbon, boron, and nitrogen:

S.N	System	Cohesive energy (eV/atom)
1.	Pristine hBN	− 6.91[1], -7.59[2,3]
2.	graphene	-7.90 [4]
3.	α, β, γ-graphyne	6.93, 7.01, 7.21 [5]
4.	Borophene	-5.99 [6]
5.	Porous borophene-graphene(pBGH)	-5.59 [7]
6.	BC_2N	−6.40 [8]
7.	C_6N_7	-6.26 [9,10]
8.	g-C_2N	-6.83 [11]
9.	T-C_3N_2	-6.62 [12]
10.	C_3N_4	−6.09 [13]

11.	Be_2C	-5.33 [14]
12.	BC_2As	-5.77 [8]
13.	**2DPA-1 (present work)**	**-5.76**
14.	**BN-2DPA (present work)**	**-5.72**
15.	**NN-2DPA (present work)**	**-5.55**
16.	**CC-2DPA (present work)**	**-5.76**

Table S3: Comparison of workfunctions of our system with other 2D systems.

S.N.	System	Workfunction (used functional) (eV)
1.	**2DPA-1 (present work)**	6.18 (PBE), 7.46 (HSE06)
2.	**BN-2DPA (present work)**	6.07 (PBE), 7.19 (HSE06)
3.	**NN-2DPA (present work)**	4.72 (PBE), 5.61 (HSE06)
4.	Graphene	4.23[15], 4.49[16], 4.6[17]
5.	Reduced graphene oxide (rGO)	5 to 5.5eV [18]
6.	Graphyne	4.06 [19], 4.04 [20]
7.	Holey graphyne	5.2[21]
8.	$g\text{-}C_3N_4$	4.29 [22]
9.	$g\text{-}C_6N_7$	6.2 [10]
10.	h-BN	5.99 [23]

Table S4: Comparison of Young-modulus of our system with other 2D systems.

S.N.	System	Young-modulus
1.	**2DPA-1 (present work)**	40.166 N/m ~ 11.76 GPa
2.	**BN-2DPA (present work)**	17.132 N/m
3.	**NN-2DPA (present work)**	27.577 N/m
4.	2DPA-1 (experimental report)	12.7±3.8 GPa [24]
5.	Polyamide/ melamine cyanurate/nanoclay composites	2.7-6.8 GPa [25]
6.	polymethylmethacrylate (acrylic), polyamide (nylon), low density polyethylene	1.99, 1.06, 0.29 GPa [26]
7.	Cu_4Se_4	55.73 N/m [27]
8.	hydrogenated Pt_2N_4	221 N/m [28]

9.	V_2S_2O	144.5 N/m [29]
10.	g-C_3N_4	187 N/m [30]
11.	Biphenylene	259.7 N/m [31]
12.	BN-biphenylene	235 N/m [32]
13.	Graphene	1000 GPa [33] or 342 N/m [34]

************************* Data Availability *****************************

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