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Supporting information for article:

Structural insight from intermolecular interaction and energy framework analysis of 2-substituted 6,7,8,9-tetrahydro-11*H*-pyrido[2,1-b]quinazolin-11-ones

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Table S1. Percentage contributions of interatomic contacts to the Hirshfeld surface for **2**, **3** and **4**.

2		**3**		**4**	
Contact	% contribution	Contact	% contribution	Contact	% contribution
H···H	51.3	H···H	30.8	H···H	47.3
C··H/H···C	22.5	O···H/H···O	29.2	C···H/H···C	21.8
N···H/H··N	12.5	C···H/H···C	18.7	O···H/H···O	12.7
O···H/H···O	9.5	H···N-N···H	8.6	N···H/H···N	7.0
C···O/O···C	2.0	C···O/O···C	4.1	C···C	5.2
N···O/O···N	1.0	N···O/O···N	2.9	C···N/N···C	2.6
C···C	0.8	O···O	2.7	C···O/O···C	2.6
C···N/N···C	0.4	C···N/N···C	1.5	N···O/O···N	0.5
		C···C	1.0	N···N	0.3
		N···N	0.5		

Table S2. Energy framework detail of interaction for 2 with symmetry operations (symmop) and distances between molecular centroids (*R*) in Å.

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	1	x, y, z	9.63	B3LYP/6-31G(d,p)	-4.6	-1.1	-15.6	8.5	-14.0
	0	-x, -y, -z	12.39	B3LYP/6-31G(d,p)	6.5	-1.2	-6.8	4.0	2.5
	0	-x+1/2, y+1/2, -z+1/2	7.92	B3LYP/6-31G(d,p)	-19.0	-4.4	-16.7	17.5	-27.0
	1	x+1/2, -y+1/2, z+1/2	7.19	B3LYP/6-31G(d,p)	-31.0	-8.0	-23.2	32.9	-38.6
	0	-x, -y, -z	4.36	B3LYP/6-31G(d,p)	-3.0	-4.3	-54.4	24.9	-38.5
	2	-x+1/2, y+1/2, -z+1/2	6.47	B3LYP/6-31G(d,p)	-8.0	-1.5	-33.3	17.0	-28.1
	1	-x, -y, -z	10.48	B3LYP/6-31G(d,p)	-1.4	-0.5	-11.6	6.4	-8.0
	0	-x, -y, -z	8.68	B3LYP/6-31G(d,p)	1.9	-0.2	-3.2	0.1	-0.8
	1	x+1/2, -y+1/2, z+1/2	8.42	B3LYP/6-31G(d,p)	-8.3	-2.9	-10.0	9.6	-13.6
	1	-x, -y, -z	11.31	B3LYP/6-31G(d,p)	-0.5	-0.1	-2.1	0.0	-2.3
		Total			**-67.4**	**-24.2**	**-176.9**	**120.9**	**-168.4**

Table S3. Energy framework detail of interaction for **3** with symmetry operations (symmop) and distances between molecular centroids (*R*) in Å.

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	2	-x+1/2, y+1/2, -z+1/2	5.38	B3LYP/6-31G(d,p)	-22.6	-5.9	-42.9	32.1	-45.8
	1	-x, -y, -z	13.29	B3LYP/6-31G(d,p)	4.0	-0.6	-5.8	1.5	-0.3
	1	-x, -y, -z	14.58	B3LYP/6-31G(d,p)	2.5	-0.2	-1.1	0.0	1.5
	2	x+1/2, -y+1/2, z+1/2	11.82	B3LYP/6-31G(d,p)	-8.1	-1.9	-5.7	4.7	-12.1
	2	-x+1/2, y+1/2, -z+1/2	10.03	B3LYP/6-31G(d,p)	-12.2	-2.8	-12.9	13.4	-17.9
	1	-x, -y, -z	4.86	B3LYP/6-31G(d,p)	-8.7	-2.6	-54.4	30.5	-39.6
	2	x, y, z	5.64	B3LYP/6-31G(d,p)	4.2	-2.4	-32.0	15.9	-15.4
	2	x+1/2, -y+1/2, z+1/2	11.64	B3LYP/6-31G(d,p)	-7.3	-2.2	-8.9	5.8	-13.6
	1	-x, -y, -z	7.16	B3LYP/6-31G(d,p)	-18.6	-4.1	-22.5	29.9	-23.8
		Total			**-66.8**	**-22.7**	**-186.2**	**133.8**	**-167.0**

Table S4. Energy framework detail of interaction for **4** with symmetry operations (symmop) and distances between molecular centroids (*R*) in Å.

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	1	-x+1/2, y+1/2, -z+1/2	10.97	B3LYP/6-31G(d,p)	0.5	-0.0	-0.6	0.0	0.0
	1	x+1/2, -y+1/2, z+1/2	14.51	B3LYP/6-31G(d,p)	-0.2	-0.0	-0.1	0.0	-0.3
	0	-x, -y, -z	3.88	B3LYP/6-31G(d,p)	-16.2	-8.1	-106.8	66.1	-75.3
	0	-x+1/2, y+1/2, -z+1/2	16.79	B3LYP/6-31G(d,p)	-0.0	-0.0	-0.2	0.0	-0.2
	0	x+1/2, -y+1/2, z+1/2	13.76	B3LYP/6-31G(d,p)	-1.9	-0.2	-1.6	0.0	-3.5
	0	x+1/2, -y+1/2, z+1/2	13.99	B3LYP/6-31G(d,p)	0.3	-0.0	-0.1	0.0	0.2
	1	x+1/2, -y+1/2, z+1/2	14.30	B3LYP/6-31G(d,p)	0.2	-0.0	-0.8	0.0	-0.5
	0	-x+1/2, y+1/2, -z+1/2	11.86	B3LYP/6-31G(d,p)	-9.1	-2.0	-19.9	16.0	-18.5
	1	x, y, z	10.65	B3LYP/6-31G(d,p)	-0.9	-0.2	-2.6	0.0	-3.4
	1	-x, -y, -z	10.10	B3LYP/6-31G(d,p)	-15.5	-2.9	-56.8	39.7	-43.4
	1	-	10.93	B3LYP/6-31G(d,p)	0.0	0.0	0.0	0.0	0.0
	0	-	14.59	B3LYP/6-31G(d,p)	-16.2	-8.1	-106.8	66.1	-75.3
	1	-	7.61	B3LYP/6-31G(d,p)	0.5	-0.0	-0.6	0.0	0.0
	0	-	7.25	B3LYP/6-31G(d,p)	0.3	-0.0	-0.1	0.0	0.2
	1	-	9.68	B3LYP/6-31G(d,p)	-0.9	-0.2	-2.6	0.0	-3.4
	0	-	12.67	B3LYP/6-31G(d,p)	-15.5	-2.9	-56.8	39.7	-43.4
	0	-	5.86	B3LYP/6-31G(d,p)	0.5	-0.0	-0.6	0.0	0.0
	1	-	6.64	B3LYP/6-31G(d,p)	-0.2	-0.0	-0.1	0.0	-0.3
	0	-	15.00	B3LYP/6-31G(d,p)	0.0	-0.0	-0.0	0.0	0.0
	1	-	11.15	B3LYP/6-31G(d,p)	-0.0	-0.0	-0.0	0.0	-0.0
	1	-	11.20	B3LYP/6-31G(d,p)	0.1	-0.0	-0.0	0.0	0.1
	0	-	12.11	B3LYP/6-31G(d,p)	-0.0	-0.0	-0.0	0.0	-0.0
	0	-	10.09	B3LYP/6-31G(d,p)	0.1	-0.0	-0.0	0.0	0.1
	0	-	11.75	B3LYP/6-31G(d,p)	-0.1	-0.0	-0.0	0.0	-0.1
		Total			**-74.2**	**-24.6**	**-357.1**	**227.6**	**-267**

Table S5. Energies of the highest occupied (HOMO) and lowest unoccupied (LUMO) molecular orbitals of the studied compounds, as obtained *in vacuo* at the B3LYP-D3BJ/6-311++G(d,p) level of theory.

Compound	E_{HOMO} [a.u.]	E_{LUMO} [a.u.]	ΔE [a.u.]	ΔE [eV]
1	-0.23338	-0.05163	0.18175	4.95
2	-0.20486	-0.04791	0.15695	4.27
3	-0.25814	-0.10641	0.15173	4.13
4 A	-0.21968	-0.06272	0.15696	4.27
4 B	-0.21766	-0.06089	0.15677	4.27

Note: $\Delta E = E_{LUMO} - E_{HOMO}$, 1 a.u. = 27.2114 eV

Table S6. Energies of the HOMO and LUMO as obtained using DMSO as implicit solvation model at the B3LYP-D3BJ/6-311++G(d,p) level of theory.

Compound	E_{HOMO} [a.u.]	E_{LUMO} [a.u.]	ΔE [a.u.]	ΔE [eV]
1	-0.23996	-0.05962	0.18034	4.91
2	-0.21129	-0.05727	0.15402	4.19
3	-0.25606	-0.11672	0.13934	3.79
4 A	-0.22665	-0.06714	0.15951	4.34
4 B	-0.22657	-0.06631	0.16026	4.36

Note: $\Delta E = E_{LUMO} - E_{HOMO}$, 1 a.u. = 27.2114 eV

Table S7. Energies of the HOMO and LUMO as obtained using methanol as implicit solvation model at the B3LYP-D3BJ/6-311++G(d,p) level of theory.

Compound	E_{HOMO} [a.u.]	E_{LUMO} [a.u.]	ΔE [a.u.]	ΔE [eV]
1	-0.23984	-0.05948	0.18036	4.91
2	-0.21116	-0.05710	0.15406	4.19
3	-0.25610	-0.11656	0.13954	3.80
4 A	-0.22653	-0.06703	0.15950	4.34
4 B	-0.22642	-0.06617	0.16025	4.36

Note: $\Delta E = E_{LUMO} - E_{HOMO}$, 1 a.u. = 27.2114 eV

Table S8. Cartesian coordinates of compound **1** (6,7,8,9-Tetra-hydro-11H-pyrido[2,1-b]quinazolin-11-one) optimized at the B3LYP-D3BJ/6-311++G(d.p) level of theory.

Atome	a / Å	b / Å	c / Å	NPA charges
C	-0.99123	-0.88254	-0.01845	0.49472
C	0.12356	1.29120	0.01977	-0.17258
C	1.36813	0.53042	-0.00179	0.17372
C	1.31047	-0.87667	-0.00637	-0.20279
C	2.60460	1.19376	0.00119	-0.17571
C	3.77745	0.46238	-0.00540	-0.21007
C	3.72670	-0.94279	-0.00986	-0.14857
C	2.51440	-1.60653	-0.00905	-0.42794
C	-2.28134	-1.66783	-0.08471	-0.37905
C	-3.51943	-0.89569	0.36071	-0.39367
C	-3.52849	0.45381	-0.34963	-0.18180
C	-2.31183	1.26816	0.05646	0.67331
H	2.60845	2.27645	0.00906	0.22537
H	4.73524	0.96858	-0.00560	0.20588
H	4.64957	-1.51170	-0.01354	0.20715
H	2.45725	-2.68789	-0.01177	0.22868
H	-2.12481	-2.58013	0.49132	0.21357
H	-2.40576	-1.98046	-1.12880	0.23807
H	-3.50297	-0.74256	1.44587	0.20332
H	-4.41889	-1.47203	0.13208	0.20405
H	-4.42235	1.03337	-0.10554	0.20462
H	-3.53168	0.29837	-1.43434	0.20934
H	-2.41758	1.62749	1.08517	0.24558
H	-2.18246	2.14952	-0.56909	0.19232
N	0.11159	-1.56052	-0.00636	-0.48438
N	-1.04669	0.50052	-0.01924	-0.51804
O	0.04296	2.50935	0.07089	-0.62499

Table S9. Cartesian coordinates of compound **2** (2-Amino-6,7,8,9-tetrahydro-11H-pyrido[2,1-b]quinazolin-11-one) optimized at the B3LYP-D3BJ/6-311++G(d.p) level of theory.

Atome	a / Å	b / Å	c / Å	NPA charges
C	-2.31038	0.94745	-0.00617	-0.21081
C	-3.44489	0.14448	-0.00810	0.16189
C	-3.28096	-1.26043	-0.00848	-0.21603
C	-2.02753	-1.83355	-0.00407	-0.17988
C	-0.87183	-1.02942	-0.00721	0.14381
C	2.76892	-1.55732	-0.09248	-0.42277
C	1.42290	-0.87301	-0.02025	0.48178
C	3.94225	-0.69648	0.36564	-0.38212
C	3.85365	0.65306	-0.33923	-0.39343
C	2.57702	1.37299	0.06200	-0.16929
C	0.15121	1.21560	0.01452	0.66854
C	-1.03690	0.36711	-0.00915	-0.14688
H	-4.79931	1.65895	0.22328	0.37460
H	-5.46798	0.12317	0.30465	0.37126

H	-2.38413	2.02859	-0.00331	0.22472
H	-4.16228	-1.89372	-0.01688	0.20106
H	-1.90475	-2.90957	-0.00174	0.22336
H	2.92354	-1.84793	-1.13910	0.21726
H	2.68267	-2.48480	0.47411	0.23420
H	3.90679	-0.55001	1.45138	0.19377
H	4.88476	-1.20140	0.14079	0.20665
H	4.70025	1.29723	-0.08812	0.21246
H	3.87467	0.50219	-1.42445	0.19581
H	2.65141	1.73608	1.09235	0.19768
H	2.38705	2.24492	-0.56140	0.22904
N	-4.72539	0.69565	-0.06827	-0.78764
N	0.37240	-1.62645	-0.00581	-0.51537
N	1.37284	0.51476	-0.02250	-0.48276
O	0.13845	2.43823	0.06779	-0.63090

Table S10. Cartesian coordinates of compound **3** (2-Nitro-6,7,8,9- tetrahydro-11H-pyrido[2,1-b]quinazolin-11-one) optimized at the B3LYP-D3BJ/6-311++G(d.p) level of theory.

Atome	a / Å	b / Å	c / Å	NPA charges
C	-1.77345	0.74945	-0.00782	-0.12537
C	-2.82341	-0.14605	0.00349	0.05994
C	-2.61880	-1.53494	0.01243	-0.16613
C	-1.33245	-2.02682	0.01189	-0.19407
C	-0.23087	-1.14450	0.00490	0.20210
C	3.42627	-1.46828	0.07073	-0.42741
C	2.04991	-0.84889	0.01381	0.51438
C	4.56146	-0.54185	-0.35383	-0.38354
C	4.39443	0.79214	0.36611	-0.39552
C	3.08890	1.44975	-0.04634	-0.17049
C	0.66792	1.16286	-0.02786	0.67441
C	-0.46942	0.24576	-0.00476	-0.17145
H	-1.94558	1.81647	-0.01863	0.26403
H	-3.47671	-2.19238	0.01931	0.24448
H	-1.13674	-3.09120	0.01807	0.23357
H	3.38418	-2.38255	-0.52189	0.23945
H	3.58091	-1.78440	1.10966	0.22570
H	5.52292	-1.00331	-0.11856	0.21141
H	4.53829	-0.38343	-1.43788	0.19650
H	5.20963	1.48148	0.13361	0.21734
H	4.40831	0.63129	1.44980	0.19819
H	2.84193	2.30171	0.58440	0.23173
H	3.15325	1.82599	-1.07191	0.20259
N	-4.20374	0.36856	0.00450	0.48370
N	1.03958	-1.66594	0.00438	-0.52742
N	1.93002	0.52496	0.01382	-0.47420
O	-5.11410	-0.45311	0.01088	-0.38434
O	-4.36130	1.58274	-0.00088	-0.37667
O	0.58985	2.37731	-0.08018	-0.60278

Table S11. Cartesian coordinates of compound **4** (N-(11-oxo-6,8,9,11-tetra¬hydro-7H-pyrido[2,1-b] quinazolin-2-yl)benzamide), conformer **A**, optimized at the B3LYP-D3BJ/6-311++G(d.p) level of theory.

Atome	a / Å	b / Å	c / Å	NPA charges
C	-0.08984	0.47820	0.05644	-0.17325
C	0.88919	-0.50248	-0.04053	0.14799
C	0.50520	-1.85620	-0.17351	-0.20615
C	-5.51243	-1.10915	-0.19114	-0.42411
C	-6.52653	-0.10928	0.35598	-0.38244
C	-6.21014	1.26547	-0.22358	-0.39380
C	-4.83259	1.72106	0.22722	-0.16933
C	-1.44227	0.13010	0.01647	-0.16257
C	-2.46678	1.16245	0.13340	0.67117
C	3.37844	-0.84574	-0.00288	0.66753
C	4.65452	-0.05474	-0.01194	-0.13269
C	4.76199	1.22727	-0.56239	-0.19418
C	5.98319	1.89615	-0.55261	-0.19866
C	7.10509	1.29169	0.01001	-0.18529
C	7.00617	0.00953	0.54987	-0.19649
C	5.79025	-0.66363	0.53077	-0.15736
C	-0.83278	-2.19477	-0.20826	-0.16849
C	-1.83636	-1.21370	-0.11551	0.15548
C	-4.07085	-0.66877	-0.07888	0.49054
H	2.36148	0.91066	0.09019	0.39296
H	0.16615	1.52642	0.16478	0.22595
H	1.26988	-2.61388	-0.23952	0.25079
H	-5.58248	-2.08467	0.29060	0.23616
H	-5.70986	-1.27704	-1.25713	0.21960
H	-7.53897	-0.42689	0.09608	0.20800
H	-6.47247	-0.07128	1.45003	0.19458
H	-6.25201	1.22153	-1.31771	0.19635
H	-6.93745	2.01622	0.09566	0.21361
H	-4.49582	2.60218	-0.31600	0.22922
H	-4.84795	1.99378	1.28741	0.19887
H	3.90956	1.69907	-1.03836	0.20192
H	6.05972	2.88343	-0.99256	0.20817
H	8.05450	1.81418	0.02007	0.20738
H	7.87879	-0.46600	0.98199	0.20867
H	5.69757	-1.66568	0.92965	0.22986
H	-1.13605	-3.22948	-0.30850	0.22660
N	2.23251	-0.08405	-0.01132	-0.59282
N	-3.16324	-1.58792	-0.15332	-0.51643
N	-3.78943	0.68371	0.04964	-0.48155
O	3.38232	-2.06649	0.02375	-0.61678
O	-2.24536	2.35455	0.29559	-0.62875

Table S12. Cartesian coordinates of compound **4** (N-(11-oxo-6,8,9,11-tetra¬hydro-7H-pyrido[2,1-b] quinazolin-2-yl)benzamide), conformer **B**, optimized at the B3LYP-D3BJ/6-311++G(d.p) level of theory.

Atome	a / Å	b / Å	c / Å	NPA charges
C	0.00437	-0.20253	0.02540	-0.17829
C	4.52466	-0.29908	0.01586	-0.13926
C	0.91142	0.84389	-0.08352	0.16283
C	5.13660	0.82048	0.59038	-0.19580
C	0.43146	2.16659	-0.21710	-0.20768
C	6.52493	0.92289	0.62442	-0.19965
C	-0.91761	2.43881	-0.24188	-0.18692
C	7.31245	-0.09007	0.08187	-0.18584
C	-1.85321	1.39204	-0.13637	0.15630
C	6.70782	-1.21363	-0.48137	-0.20369
C	-5.51591	1.06801	-0.18642	-0.42377
C	-4.04977	0.71494	-0.08258	0.49254
C	5.32226	-1.32212	-0.50583	-0.14165
C	-6.46428	0.00945	0.36804	-0.38239
C	-6.06663	-1.34481	-0.20964	-0.39385
C	-4.66123	-1.71311	0.23479	-0.16989
C	-2.33201	-1.01358	0.13004	0.67090
C	3.03625	-0.49004	-0.04104	0.66807
C	-1.36900	0.07859	-0.00316	-0.14677
H	2.84030	1.53064	-0.14459	0.38975
H	0.33673	-1.22330	0.12592	0.26865
H	4.54131	1.60014	1.05262	0.20053
H	1.14299	2.98250	-0.30206	0.19893
H	6.99023	1.78712	1.08362	0.20733
H	-1.28496	3.45206	-0.34425	0.22548
H	8.39286	-0.00862	0.10632	0.20712
H	7.31787	-2.00693	-0.89711	0.20874
H	-5.72930	1.22214	-1.25141	0.21933
H	-5.64174	2.03829	0.29471	0.23502
H	4.83481	-2.19427	-0.92252	0.22987
H	-6.40247	-0.02222	1.46189	0.19443
H	-7.49563	0.26379	0.11261	0.20741
H	-6.74467	-2.13801	0.11531	0.21356
H	-6.11680	-1.30665	-1.30366	0.19617
H	-4.65355	-1.98400	1.29553	0.19920
H	-4.27321	-2.57319	-0.30795	0.23031
N	2.30817	0.67933	-0.05653	-0.60258
N	-3.19965	1.68793	-0.16586	-0.52248
N	-3.68542	-0.61386	0.04880	-0.48357
O	2.53266	-1.59939	-0.08445	-0.60488
O	-2.04665	-2.18733	0.30306	-0.61347

Table S13. Quantum chemical data obtained through optimization of the condensed phase conformers at the B3LYP-D3BJ/6-311G(d,p) level of theory.

Compound	A[a]	B	C	E	ZPE	$\|\mu_a\|$	$\|\mu_b\|$	$\|\mu_c\|$	$\|\mu\|$
1	1.426	0.442	0.342	-649.501144	-649.279494	2.23	1.60	0.13	2.75
2	1.373	0.336	0.273	-704.881533	-704.643404	0.63	1.06	0.82	1.48
3	1.215	0.225	0.192	-854.070005	-853.846291	8.16	2.65	0.14	8.58
4A	0.841	0.086	0.079	-1049.397206	-1049.067866	1.89	2.41	0.16	3.06
4B	0.879	0.088	0.081	-1049.395105	-1049.065930	0.56	3.62	0.23	3.67

[a] rotational constants *A. B. C* in GHz; electronic energies *E* in Hartree; sum of electronic and zero-point energies *ZPE* in Hartree; dipole moment components in the principle axis of inertia system; dipole moment μ in Debye.

Table S14. Quantum chemical data obtained through optimization of the condensed phase conformers at the B3LYP/6-311G(d,p) level of theory.

Compound	A[a]	B	C	E	ZPE	$\|\mu_a\|$	$\|\mu_b\|$	$\|\mu_c\|$	$\|\mu\|$
1	1.423	0.441	0.341	-649.443915	-649.222621	2.24	1.61	0.12	2.76
2	1.371	0.335	0.272	-704.820328	-704.582589	0.63	1.06	0.82	1.47
3	1.213	0.225	0.191	-854.006712	-853.783384	8.17	2.65	0.13	8.59
4A	0.840	0.085	0.078	-1049.305655	-1048.976867	1.92	2.40	0.16	3.08
4B	0.879	0.087	0.080	-1049.303500	-1048.974872	0.60	3.63	0.23	3.68

[a] rotational constants *A. B. C* in GHz; electronic energies *E* in Hartree; sum of electronic and zero-point energies *ZPE* in Hartree; dipole moment components in the principle axis of inertia system; dipole moment μ in Debye.

Table S15. Quantum chemical data obtained through optimization of the condensed phase conformers at the B3LYP-D3/6-311G(d,p) level of theory.

Compound	A[a]	B	C	E	ZPE	$\|\mu_a\|$	$\|\mu_b\|$	$\|\mu_c\|$	$\|\mu\|$
1	1.423	0.441	0.342	-649.467704	-649.246163	2.24	1.60	0.14	2.76
2	1.370	0.335	0.272	-704.846297	-704.608244	0.63	1.05	0.80	1.47
3	1.213	0.225	0.191	-854.034282	-853.810645	8.18	2.65	0.15	8.60
4A	0.839	0.085	0.078	-1049.344319	-1049.015183	1.93	2.41	0.21	3.10
4B	0.878	0.088	0.081	-1049.342243	-1049.013273	0.61	3.62	0.21	3.68

[a] rotational constants *A. B. C* in GHz; electronic energies *E* in Hartree; sum of electronic and zero-point energies *ZPE* in Hartree; dipole moment components in the principle axis of inertia system; dipole moment μ in Debye.

Table S16. Quantum chemical data obtained through optimization of the condensed phase conformers at the PBE0-D3/6-311G(d,p) level of theory.

Compound	A[a]	B	C	E	ZPE	$\|\mu_a\|$	$\|\mu_b\|$	$\|\mu_c\|$	$\|\mu\|$
1	1.435	0.446	0.345	-648.704896	-648.481605	2.29	1.58	0.14	2.79
2	1.381	0.338	0.275	-704.023101	-703.783086	0.65	1.01	0.81	1.45
3	1.223	0.227	0.193	-853.065292	-852.839511	8.06	2.60	0.16	8.47
4A	0.845	0.086	0.079	-1048.117595	-1047.785757	1.95	2.45	0.17	3.13
4B	0.884	0.089	0.081	-1048.115524	-1047.783845	0.62	3.57	0.25	3.63

[a] rotational constants *A. B. C* in GHz; electronic energies *E* in Hartree; sum of electronic and zero-point energies *ZPE* in Hartree; dipole moment components in the principle axis of inertia system; dipole moment μ in Debye.

Table S17. Quantum chemical data obtained through optimization of the condensed phase conformers at the B3LYP-D3BJ/6-311G(d,p) level of theory.

Compound	A[a]	B	C	$\|\mu_a\|$	$\|\mu_b\|$	$\|\mu_c\|$	$\|\mu\|$	E	Erel	ZPE
[b]1 - twist 1	1.4258	0.4421	0.3423	-2.23	-1.60	-0.13	2.75	-649.501144	0.00	-649.279494
1 - twist 2	1.4257	0.4421	0.3423	-2.23	-1.60	0.13	2.75	-649.501144	0.00	-649.279494
1 - boat 1	1.3756	0.4526	0.3536	2.16	-1.60	0.03	2.69	-649.499932	3.18	-649.278184
1 - boat 2	1.3757	0.4526	0.3536	-2.16	-1.60	0.03	2.69	-649.499932	3.18	-649.278183
2 - twist 1	1.3726	0.3357	0.2729	0.62	-1.05	1.05	1.61	-704.881509	0.06	-704.643391
[b]2 - twist 2	1.3727	0.3357	0.2729	0.63	-1.06	0.82	1.48	-704.881533	0.00	-704.643404
2 - boat 1	1.3230	0.3428	0.2810	0.65	-1.05	1.14	1.68	-704.880379	3.03	-704.642164
2 - boat 2	1.3218	0.3429	0.2811	-0.49	-1.06	-0.71	1.37	-704.880392	3.00	-704.642175
[b]3 - twist 1	1.2145	0.2254	0.1917	-8.16	-2.65	-0.14	8.58	-854.070005	0.00	-853.846291
3 - twist 2	1.2145	0.2254	0.1917	-8.16	-2.65	0.14	8.58	-854.070005	0.00	-853.846290
3 - boat 1	1.1724	0.2295	0.1964	8.04	-2.64	-0.18	8.47	-854.068625	3.62	-853.844819
3 - boat 2	1.1725	0.2295	0.1964	-8.04	-2.64	-0.18	8.47	-854.068625	3.62	-853.844819
[b]4 A - twist 1	0.8407	0.0857	0.0786	1.89	2.41	0.16	3.06	-1049.397206	0.03	-1049.067866
4 A - twist 2	0.8408	0.0857	0.0786	1.89	2.39	-0.05	3.04	-1049.397218	0.00	-1049.067870
4 A - boat 1	0.8186	0.0867	0.0797	1.80	2.42	0.14	3.02	-1049.396045	3.08	-1049.066592
4 A - boat 2	0.8183	0.0866	0.0798	-1.78	2.37	0.07	2.96	-1049.396051	3.06	-1049.066588
4 B - twist 1	0.8791	0.0879	0.0807	-0.56	3.65	0.03	3.69	-1049.395095	5.57	-1049.065921
[b]4 B - twist 2	0.8790	0.0879	0.0808	-0.56	3.62	-0.23	3.67	-1049.395105	5.55	-1049.065930
4 B - boat 1	0.8550	0.0888	0.0820	-0.48	3.67	0.29	3.71	-1049.393878	8.77	-1049.064603
4 B - boat 2	0.8550	0.0888	0.0820	0.48	3.67	0.29	3.71	-1049.393878	8.77	-1049.064603

[a] rotational constants *A. B. C* in GHz; electronic energies *E* in Hartree; sum of electronic and zero-point energies *ZPE* in Hartree; dipole moment components in the principle axis of inertia system; dipole moment μ in Debye.

[b] conformers corresponding to the observed X-ray structures (see also Table 6 in the manuscript).

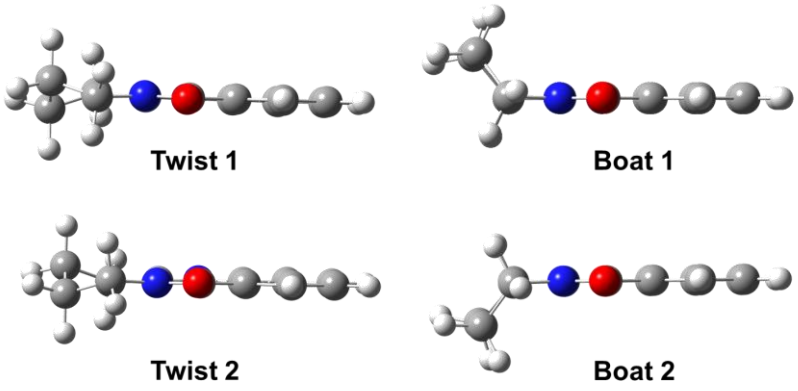

Figure S1. Structures of the four different ring conformations (view along the OC carbonyl bond) that can be adopted by the pyridine ring in compound **1** (6,7,8,9-Tetra-hydro-11H-pyrido[2,1-b]quinazolin-11-one). This referencing is used throughout the main text and in Table S17 above to differentiate between the conformers. Note that within the crystal structures either a twist 1 or twist 2 conformation is adopted.

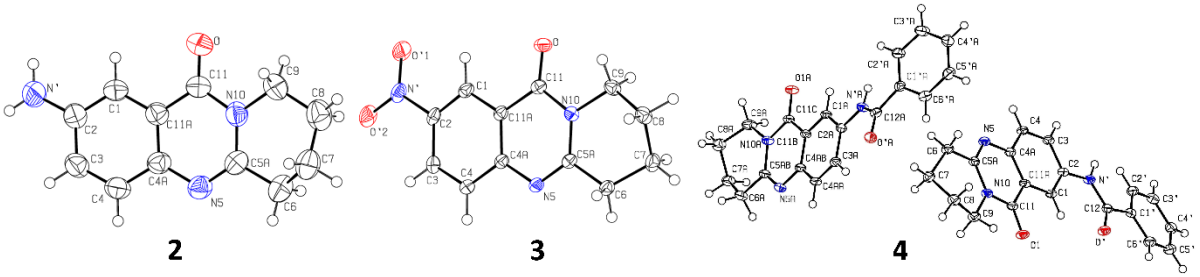

Figure S2. Displacement ellipsoid plot of compound **2** (298 K, ellipsoids are drawn at 30% probability); of compounds **3** and **4** (100 K, ellipsoids are drawn at 50% probability) with the atom-labelling scheme. H atoms are shown as spheres of arbitrary radius.

Figure S3. Overlay diagram for compounds **1** (red), **2** (blue), **3** (yellow) and **4** (pink).

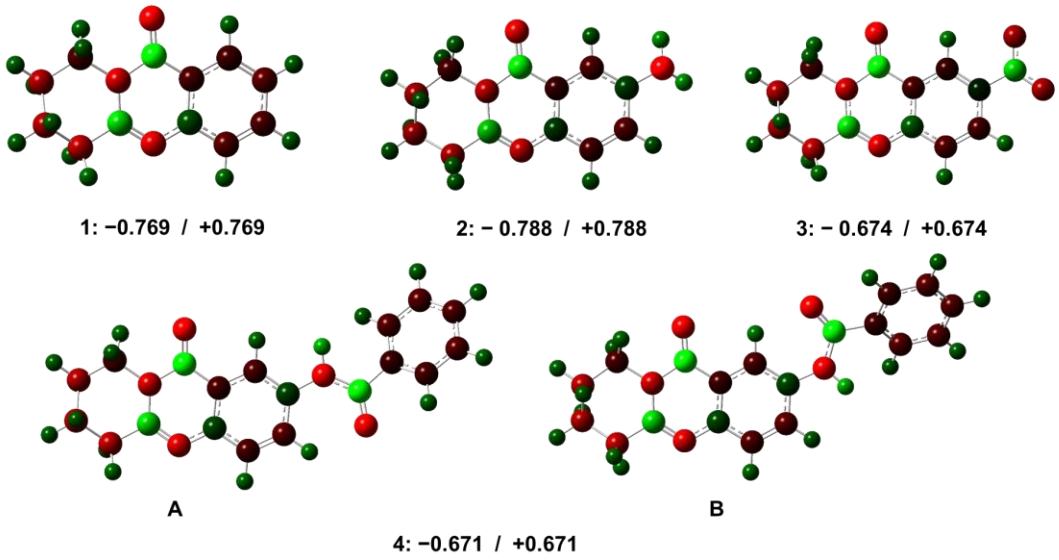

Figure S4. Natural population charge analysis of the 4 investigated compounds (1, 2, 3, 4). The absolute values of the atomic charges for each atom are given with the Cartesian coordinates in Tables S8-S12.

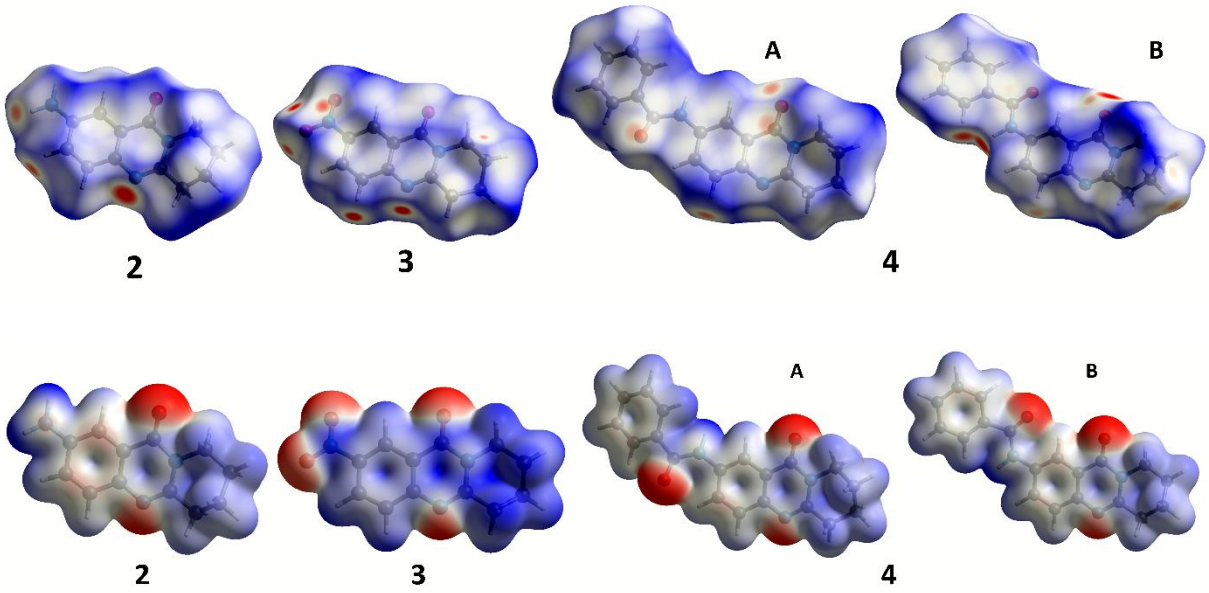

Figure S5. Views of the Hirshfeld surfaces (top) over dnorm, and electrostatic potential (bottom).