Supplementary material for the manuscript

The Whole Range of Hydrogen Bonds in One Crystal Structure: Neutron Diffraction and Charge Density Studies of *N,N*-Dimethylbiguanidinium Bis(hydrogensquarate)

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a) Compilation of neutron diffraction studies on intermolecular O-H···O bonds with D···A < 2.6 Å

b) Compilation of neutron diffraction studies on intramolecular O-H···O bonds with D···A < 2.6 Å

c) Diagrams concerning the data quality of the X-ray diffraction experiment on **1**

d) Beam stability in the neutron diffraction experiment on **1**

e) Table S1. Completeness of X-ray and neutron intensity data for **1**

f) Scatterplot F^2_{obs} vs F^2_{calc} for X-ray refinement result on **1**

g) Scatterplot F^2_{obs} vs F^2_{calc} for neutron refinement result on **1**

h) Isosurface maps showing the electrostatic potentials of a dication and two H-bonded monoanions in the crystal structure of **1**

i) Table S2. Interatomic distances, bond paths R_{ij}, d_1, d_2 (Å), electron densities ρ (eÅ^{-3}) and Laplacian of the electron density $\nabla^2\rho$ (eÅ^{-5}) in the bond critical points for covalent bonds in **1**

j) Table S3. Interatomic distances, bond paths R_{ij}(Å), electron densities ρ (eÅ^{-3}), Laplacian of the electron density $\nabla^2\rho$ (eÅ^{-5}), kinetic (*G, au*) and potential energy density *(V, au*) in the bond critical points for hydrogen bonds in **1**

a) Compilation of neutron diffraction studies on intermolecular O-H···O bonds with D···A < 2.6 Å.

The search is based on the CSD data base, Version 5.32, including the updates of Feb. 2011.

	O-H···O distance [Å]	CSD refcode	Reference
1.	2.4 2.567 2.405 2.575 2.417 2.593 2.418 2.568 2.421 2.580 2.423 2.562 2.423 2.570 2.425 2.561 2.429 2.554 2.430 2.568 2.433 2.571 2.403 2.573	CRBAMP05 CRBAMP06 CRBAMP07 CRBAMP17 CRBAMP09 CRBAMP12 CRBAMP13 CRBAMP16 CRBAMP18 CRBAMP14 CRBAMP15 CRBAMP08	Wilson C.C. (2001). "Migration of the proton in the strong O-HO hydrogen bond in urea-phosphoric acid (1/1)". Acta Crystallogr. Sect.B 57:435-439
2.	2.403 2.582 2.411 2.583 2.417 2.581 2.418 2.590 2.419 2.585 2.420 2.586 2.420 2.586	CRBAMP20 CRBAMP19 CRBAMP23 CRBAMP22 CRBAMP25 CRBAMP24 CRBAMP21	Wilson C.C., Shankland K., Shankland N. (2001). "Single-crystal neutron diffraction of urea-phosphoric acid: evidence for H-atom migration in a short hydrogen bond between 150 K and 350 K". Z. Kristallogr. 216:303-306
3.	2.407 2.460 2.599	CBZSUL01	Roziere J., Williams J.M. (1978). "The hydrated proton $H^+(H_2O)n$. VII. A neutron diffraction study of the isolated diaquooxonium ion $H_3O^+ \cdot 2H_2O$ in

			2,5-dichlorobenzenesulphonic acid trihydrate". J. Chem. Phys. 68:2896-2901
4.	2.408 2.427 2.467 2.468 2.476 2.491	TAGTEB TAGTAX BAHOXH11 BAHOXH01 BAHOXH12	Chaix-Pluchery O., Mutin J.C., Bouillot J., Niepce J.C. (1989). "Neutron structure refinement of barium oxalate-oxalic acid dihydrate, $BaC_2O_4.H_2C_2O_4.2H_2O$ and of related nonstoichiometric hydrates". Acta Crystallogr. Sect. C 45:1699-1705
5.	2.409 2.59	CRBAMP01	Rodrigues B.L., Tellgren R., Fernandes N.G. (2001). "Experimental electron density of urea-phosphoric acid (1/1) at 100 K". Acta Crystallogr. Sect.B 57:353-358
6.	2.414	SLBZAC01	Attig R., Williams J.M. (1976). "The hydrated proton $H^+(H_2O)n$. 6. A neutron diffraction study of the isolated group $H_5O_2^+ \cdot H_2O$ in o-sulfobenzoic acid trihydrate". Inorg.Chem. 15:3057–3061
7.	2.421 2.589	CRBAMP11	Kostansek E.C., Busing W.R. (1972). "A single-crystal neutron-diffraction study of urea-phosphoric acid". Acta Crystallogr. Sect.B 28: 2454-2459
8.	2.424 2.425	XUGFAH01	Smrcok L., Havlicek D., Kaman O., Rundlof H. (2009). "1,4-diazabicyclo[2.2.2]octane-1,4-diium dihydrogen phosphate monohydrate from X-ray and neutron data". Z. Kristallogr. 224:174-178
9.	2.428	HMTOFA07	Nygren C.L., Wilson C.C., Turner J.F.C. (2005). "Electron and Nuclear Positions in the Short Hydrogen Bond in Urotropine-N-oxide·Formic Acid". J. Phys. Chem. A 109:1911–1919
10.	2.431	ETDCOH11	Roziere J., Williams J.M. (1976). "The hydrated proton H+(H2O)n. IV. A high-precision neutron diffraction study of the diaquohydrogen ion, (H2OHOH2)+, in trans-dichlorobis(ethylenediamine)cobalt(III) chloride hydrochloride dihydrate". Inorg. Chem. 15:1174-1178
11.	2.433 2.544	SATHIE01	Silvestre J.-P., Bkouche-Waksman I., Heger G., Dao N.Q. (1990). New J. Chem. 14:29-35
12.	2.437 2.476	ETHDPH01	Silvestre J.-P., Dao N.Q., Heger G., Cousson A. (2002). "Refinement by Neutron Diffraction of the crystal structure of hydroxyethylidene bisphosphonic acid monohydrate: $C(CH_3)(OH)(PO_3H_2)_2 \cdot H_2O$". Phosphorus, Sulfur, Silicon, Relat.Elem. 177:277-288
13.	2.437 2.574	JOTQOY03	Calleja M., Mason S.A., Prince P.D., Steed J.W., Wilkinson C. (2001). "Single crystal neutron and X-ray diffraction studies of $(H_7O_3)[AuCl_4]$·15-crown-5". New J. Chem. 25:1475-1478
14.	2.437 2.442 2.445 2.453	KHDCMT02 KHDCMT01	Olovsson I., Ptasiewicz-Bak H., Gustafsson T., Majerz I. (2001). "Asymmetric hydrogen bonds in centrosymmetric environment: neutron study of very short hydrogen bonds in potassium hydrogen dichloromaleate". Acta Crystallogr. Sect.B 57:311-316
15.	2.437 2.443	KHDCMT04	Olovsson I., Ptasiewicz-Bak H., Gustafsson T., Majerz I. (2002). "Asymmetric hydrogen bonds in centrosymmetric

	2.44 2.442	KHDCMT03	environment. II. Neutron study of very short hydrogen bonds in potassium hydrogen dichloromaleate at 90 K and 170 K". Acta Crystallogr. Sect.B 58:627-631
16.	2.437 2.443	KHDFRM12 KHDFRM11	Hermansson K., Tellgren R., Lehmann M.S. (1983). "Neutron diffraction studies of potassium hydrogen diformate, $KH(HCOO)_2$, at 120 and 295 K". Acta Crystallogr. Sect.C 39:1507-1510
17.	2.440 2.452	KHMTAR01	Currie M., Speakman J.C., Kanters J.A., Kroon J. (1975). "Crystal structure of the acid salts of some dibasic acids. Part IX. Potassium hydrogen *meso*-tartrate: a neutron diffraction study". J. Chem. Soc., Perkin Trans. 2:1549-1554
18.	2.443	VITLOZ01	Gao Q., Clancy L., Weber H.-P., Craven B.M., McMullan R.K. (1991). "Structure of the adipate complex $[Na_2(C_6H_9O_4)_2(C_6H_{10}O_4)]\cdot 2H_2O$ from neutron diffraction at 220 and 295 K". Acta Crystallogr. Sect.B 47:368-375
19.	2.444	KHSUCC01	McAdam A., Currie M., Speakman J.C. (1971). "The crystal structures of the acid salts of some dibasic acids. Part IV. Potassium hydrogen succinate: X-ray and neutron-diffraction studies". J. Chem. Soc. A:1994-1997
20.	2.449 2.476	KAOXYA01	Albertsson J., Grenthe I. (1973). "A neutron diffraction study of potassium and rubidium hydrogen oxydiacetate. The dynamics of their hydrogen bonds". Acta Crystallogr. Sect.B 29:2751-2760
21.	2.449	DGLYCN01	Sato S. (1968). "Crystal Structure and Phase Transition of $(Glycine)_2HNO_3$ I. Crystal Structure Determination of Ferro- and Paraelectric Phases". J. Phys. Soc. Jpn. 25:185-201
22.	2.461	WEHZAL01	Chitra R., Choudhury R.R. (2007). "Investigation of hydrogen-bond network in bis(glycinium) oxalate using single-crystal neutron diffraction and spectroscopic studies". Acta Crystallogr. Sect.B 63:497-504
23.	2.465	DETSBR01	Bideau J.-P., Bravic G., Filhol A.(1977). "Liaison hydrogène et cétoénolisation en série barbiturique à l'état cristallin. II. Structure cristalline par diffraction des neutrons de l'acide diéthyl-1,3-thio-2 barbiturique". Acta Crystallogr. Sect.B 33:3847-3849
24.	2.466 2.549 2.566 2.579 2.583 2.469 2.536 2.556 2.564 2.58	HISTPA14 HISTPA12	Mata I., Espinosa E., Molins E., Veintemillas S., Maniukiewicz W., Lecomte C., Cousson A., Paulus W. (2006). "Contributions to the application of the transferability principle and the multipolar modeling of H atoms: electron-density study of L-histidinium dihydrogen orthophosphate orthophosphoric acid. I". Acta Crystallogr. Sect. A 62:365-378
25.	2.475 2.476 2.481 2.491	HUCLEW03 HUCLEW01 HUCLEW02 HUCLEW04	Vishweshwar P., Babu N.J., Nangia A., Mason S.A., Puschmann H., Mondal R., Howard J.A.K. (2004). "Variable Temperature Neutron Diffraction Analysis of a Very Short O–H···O Hydrogen Bond in 2,3,5,6-Pyrazinetetracarboxylic

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26.	2.477 2.563	NINDOD01	Selenius C.-O., Lundgren J.-O. (1980). "Neutron diffraction study of 2-nitro-1,3-indandione dihydrate". Acta Crystallogr. Sect.B 36:3172-3175
27.	2.479	UROXAL01	van Hummel G.J., Helmholdt R.B. (1991). "The structure of urea-oxalic acid (2/1) determined by neutron diffraction at 100 K". Acta Crystallogr. Sect.C 47:213-215
28.	2.484	QADYAW	Calleja M., Mason S.A., Prince P.D., Steed J.W., Wilkinson C. (2003). "Anisotropic thermal expansion in 18-crown-6·2 H_2O·2 HNO_3". New J. Chem. 27:28-31
29.	2.486	DTBUPA01	Reis A.H., Peterson S.W., Druyan M.E., Gebert E., Mason G.W., Peppard D.F. (1976). "Sterically hindered solvent extractants. 2. A neutron-diffraction study of the di-tert-butylphosphinic acid dimer showing strong asymmetric hydrogen bonding". Inorg.Chem. 15:2748–2752
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31.	2.496	AMBACO07	Brown C.J., Ehrenberg M. (1985). "Anthranilic acid, $C_7H_7NO_2$, by neutron diffraction". Acta Crystallogr. Sect.C 41:441-443
32.	2.498	KHDCAC10	Hadzi D., Leban I., Orel B., Iwata M.,Williams J.M. (1979). "Neutron diffraction and vibrational spectroscopic study of single crystals of KH(D)($CHCl_2COO)_2$". J. Cryst. Mol. Struct. 9:117-134
33.	2.501 2.516	TGLYSU11	Kay M.I., Kleinberg R. (1973). "The crystal structure of triglycine sulfate". Ferroelectrics 5:45-52
34.	2.506	OXACDH04	Sabine T.M., Cox G.W., Craven B.M. (1969). "A neutron diffraction study of α-oxalic acid dihydrate". Acta Crystallogr. Sect.B 25:2437-2441
35.	2.512 2.566	XOBMIK01	Navaza A., Chevrier G., Kiat J.M., Barbey C. (2002). "Contribution of Neutron Diffraction to the Crystal Structure Determination of 1-Hydroxy-1-Phosphono-Pentyl-Phosphonic Acid Dimethyl Ammonium Salt". J. Solid State Chem., 167:441-445
36.	2.518	KHOXAL02	Moore F.H., Power L.F. (1971). "The crystal structure of potassium hydrogen oxalate by neutron diffraction". Inorg. Nucl. Chem. Lett. 7:873-875
37.	2.519	LGLUAC11	Lehmann M.S., Koetzle T.F., Hamilton W.C. (1972). "Precision neutron diffraction structure determination of protein and nucleic acid components. VIII: the crystal and molecular structure of the β-form of the amino acid L-glutamic acid". J. Cryst. Mol. Struct. 2:225-233
38.	2.521	TOLSAM12	Lundgren J.-O., Williams J.M. (1973). "The hydrated proton

	2.524 2.538		$H^+(H_2O)_n$. I. A single crystal neutron diffraction study of the oxonium ion in *p*-toluenesulfonic acid monohydrate, $H_3O^+CH_3C_6H_4SO_3^-$". J. Chem. Phys. 58:788-796
39.	2.522 2.579	TFMSUL02	Lundgren J.-O., Tellgren R., Olovsson I. (1978). "Neutron diffraction study of oxonium trifluoromethanesulphonate". Acta Crystallogr. Sect.B 34:2945-2947
40.	2.524	CYSTAC01	Ramanadham M., Sikka S.K., Chidambaram R. (1973). "Refinement of hydrogen-atom positions in L-cysteic acid.H_2O from neutron diffraction data". Acta Crystallogr. Sect.B 29:1167-1170
41.	2.525 2.595	PUTRDP11	Takusagawa F., Koetzle T.F. (1979). "A study of the charge density in putrescine diphosphate at 85 K". Acta Crystallogr. Sect.B 35:867-877
42.	2.527	DMAHOX01	Thomas J.O. (1977). "Hydrogen bond studies. CXXII. A neutron diffraction and *X-N* deformation-electron-density study of dimethylammonium hydrogen oxalate, $(CH_3)_2NH_2HC_2O_4$, at 298 K". Acta Crystallogr. Sect.B 33:2867-2876
43.	2.529 2.547	KHPHAL04 KHPHAL05	Harte S.M., Parkin A., Goeta A., Wilson C.C. (2005). "Using neutrons and X-rays to study the effect of temperature on the short hydrogen bond in potassium hydrogen phthalate". J. Mol. Struct. 741:93-96
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45.	2.533 2.544 2.545 2.546 2.547	KECYBU12 KECYBU16 KECYBU13	Semmingsen D., Tun Z., Nelmes R.J., McMullan R.K., Koetzle T.F. (1995). "On the temperature dependence of the hydrogen bond order in squaric acid: neutron diffraction studies at four different temperatures[1]". Z. Kristallogr. 210:934-947
46.	2.542	DLASPA02	Sequeira A., Rajagopal H., Ramanadham H. (1989). "A neutron study on the structure of DL-aspartic acid". Acta Crystallogr. Sect.C 45:906-908
47.	2.546 2.563	KHFUMA01	Gupta M.P., Prasad N. (1973). "The crystal structure of dipotassium tetrahydrogen trifumarate: $2KC_4H_3O_4 \cdot C_4H_4O_4$, a neutron-diffraction study". Z. Kristallogr. 137:173-178
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51.	2.553 2.554	KECYBU06	Semmingsen D., Hollander F.J., Koetzle T.F. (1977). "A neutron diffraction study of squaric acid (3,4-dihydroxy-3-cyclobutene-1,2-dione)". J. Chem. Phys. 66:4405-4412
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53.	2.561	AHOXLH04	Fernandes N.G., Tellgren R. (1989). "Ammonium hydrogen oxalate hemihydrate: X-ray and neutron diffraction studies". Acta Crystallogr. Sect.C 45:499-504
54.	2.562	ACYGLY11	Mackay M.F. (1975). Cryst. Struct. Commun. 4:225-228
55.	2.566	NHOXAL14	Delaplane R.G., Tellgren R., Olovsson I. (1984). "Neutron diffraction study of sodium hydrogen oxalate monohydrate, $NaHC_2O_4.H_2O$, at 120 K". Acta Crystallogr. Sect.C 40:1800-1803
56.	2.567 2.597	LARGPH04	Espinosa E., Lecomte C., Molins E., Veintemillas S., Cousson A., Paulus W. (1996). "Electron density study of a new non-linear optical material: L-arginine phosphate monohydrate (LAP). Comparison between *X*-*X* and *X*-(*X* + *N*) refinements". Acta Crystallogr. Sect.B 52:519-534
57.	2.569 2.591	DINICA11 DINICA12	Cowan J.A., Howard J.A.K., McIntyre G.J., Lo S.M.-F., Williams I.D. (2005). "Variable-temperature neutron diffraction studies of the short, strong hydrogen bonds in the crystal structure of pyridine-3,5-dicarboxylic acid". Acta Crystallogr. Sect.B 61:724-730
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61.	2.572	WIFBUI01	Adam K.R., Antolovich M., Atkinson I.M., Leong A.J., Lindoy L.F., McCool B.J., Davis R.L., Kennard C.H.L., Tasker P.A. (1994). "On the nature of the host–guest interaction between cyclam and 4-*tert*-butylbenzoic acid—a

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62.	2.574 2.593 2.596	TAPBOB06	Kazimirov V.Yu., Sarin V.A., Ritter C., Shuvalov L.A. (1999). "Neutron Diffraction Study of the Atomic Structure of the Ferroelectric Phase of $(CH_3)_2NH_2Al(SO_4)_2 \cdot 6H_2O$ Ferroelectric-Ferroelastic (DMAAS)". Kristallografiya(Russ.)(Crystallogr.Rep.) 44:56-61
63.	2.576 2.582	SOGGEA	Robl C., Kuhs W.F. (1991). "A neutron diffraction study on hydrogen bonding in the mineral mellite $(Al_2[C_6(COO)_6] \cdot 16H_2O)$ at 15 K". J. Solid State Chem. 92:101-109
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65.	2.58 2.583 2.587 2.588	CLBZAP06 CLBZAP08 CLBZAP07 CLBZAP05	Wilson C.C., Xu X., Florence A.J., Shankland N. (2006). "Temperature dependence of proton transfer in 4-chlorobenzoic acid". New J. Chem. 30:979-981
66.	2.581	LGLUAC03	Lehmann M.S., Nunes A.C. (1980). "A short hydrogen bond between near identical carboxyl groups in the α-modification of L-glutamic acid". Acta Crystallogr. Sect.B 36:1621-1625
67.	2.585	DIALAC02	Craven B.M., Sabine T.M. (1969). "The 5-hydroxyl configuration in dialuric acid monohydrate by neutron crystal structure determination". Acta Crystallogr. Sect.B 25:1970-1978
68.	2.588	NAOXAP11	Pedersen B.F., Kvick A. (1989). "Neutron diffraction study of sodium oxalate monoperhydrate at 123 K". Acta Crystallogr. Sect.C 45:1724-1727
69.	2.591	KOXPHY11	Pedersen B.F., Kvick A. (1990). "Neutron diffraction study of potassium oxalate monoperhydrate at 123 K". Acta Crystallogr. Sect.C 46:21-23
70.	2.594	BBZSUL01	Lundgren J.-O. (1979). "Neutron diffraction study of diaquaoxonium 2,5-dibromobenzenesulphonate". Acta Crystallogr. Sect.B 35:780-783
71.	2.596	UREANT02	Worsham Jnr. J.E., Busing W.R. (1969). "The crystal structure of uronium nitrate (urea nitrate) by neutron diffraction". Acta Crystallogr. Sect.B 25:572-578
72.	2.599	NUWRIG01	Bau R., Schreiber A., Metzenthin T., Lu R.S., Lutz F., Klooster W.T., Koetzle T.F., Seim H., Kleber H.-P., Brewer F., Englard S. (1997). "Neutron Diffraction Structure of (2*R*,3*R*)-L-(−)-[2-D]Carnitine Tetrachloroaurate, $[(CH_3)_3N\text{-}CH_2\text{-}CHOH\text{-}CHD\text{-}COOH]^+[AuCl_4]^-$: Determination of the Absolute Stereochemistry of the Crotonobetaine-to-Carnitine Transformation Catalyzed by L-Carnitine Dehydratase from *Escherichia coli*". J. Am. Chem. Soc., 119:12055–12060
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b) Compilation of neutron diffraction studies on intramolecular O-H···O bonds with D···A < 2.6 Å.

The search is based on the CSD data base, Version 5.32, including the updates of Feb. 2011.

	O-H···O distance [Å]	CSD refcode	Reference
1.	2.384	TEHNAV	Wozniak K., Wilson C.C., Knight K.S., Jones W., Grech E. (1996). "Neutron diffraction of a complex of 1,8-bis(dimethylamino)naphthalene with 1,2-dichloromaleic acid". Acta Crystallogr. Sect.B 52:691-696
2.	2.388 2.393	LIHPHM01	Kuppers H., Kvick A., Olovsson I. (1981). "Hydrogen bond studies. CXLII. Neutron diffraction study of the two very short hydrogen bonds in lithium hydrogen phthalate-methanol". Acta Crystallogr. Sect.B 37:1203-1207
3.	2.390	MOHNIA01	Hussain M.S., Schlemper E.O., Yelon W.B. (1981). "Neutron diffraction study of the orthorhombic form of [3,3'-dimethyl-3,3'-(2-nitropropanediylidenediamino)bis(2-butanone oximato)-*N*,*N*',*N*'',*N*''']nickel(II)". Acta Crystallogr. Sect.B 37:347-351
4.	2.390 2.429	AMECOP01	Jones D.J., Roziere J., Lehmann M.S. (1986). "Very short OHO hydrogen bonds in a binuclear transition-metal complex. Neutron-diffraction study at 120 K of $[Co_2\{H_2N(CH_2)_2OH\}_3\{H_2N(CH_2)_2O\}_3][ClO_4]_3\cdot 0.5H_2O$". J. Chem. Soc., Dalton Trans., 651-655
5.	2.391	CUHOPT04	Bartl H., Kuppers H. (1980). "Neutronenbeugungsuntersuchung der extrem kurzen Wasserstoffbrücke im Kupfer-Dihydrogen-Diphthalat-Dihydrat". Z. Kristallogr., 152:161-167
6.	2.393	IMZMAL11	Hsu B., Schlemper E.O. (1980). "*X-N* deformation density studies of the hydrogen maleate ion and the imidazolium ion". Acta Crystallogr. Sect.B 36:3017-3023
7.	2.394 2.395	AJOHOW AJOHEM	Mallinson P.R., Smith G.T., Wilson C.C., Grech E., Wozniak K. (2003). "From Weak Interactions to Covalent Bonds: A Continuum in the Complexes of 1,8-Bis(dimethylamino)-naphthalene". J. Am. Chem. Soc. 125:4259-4270
8.	2.404	CIRVAA01	Vanhouteghem F., Lenstra A.T.H., Schweiss P. (1987). "Magnesium bis(hydrogen maleate) hexahydrate, $[Mg(C_4H_3O_4)_2].6H_2O$, studied by elastic neutron diffraction and *ab initio* calculations". Acta Crystallogr. Sect.B 43:523-528
9.	2.405 2.419 2.423	IRETII02 LECFIJ	Cowan J.A., Howard J.A.K., Mason S.A., McIntyre G.J., Lo S.M.-F., Mak T., Chui S.S.-Y., Cai J., Cha J.A., Williams I.D. (2006). "Neutron diffraction studies of the 1:1 and 2:1 cocrystals of benzene-1,2,4,5-tetracarboxylic acid and 4,4'-bipyridine". Acta Crystallogr. Sect.C 62: o157-o161
10.	2.406	ACRECO01	Schultz A.J., Srinivasan K., Teller R.G., Williams J.M., Lukehart C.M. (1984). "Single-crystal, time-of-flight,

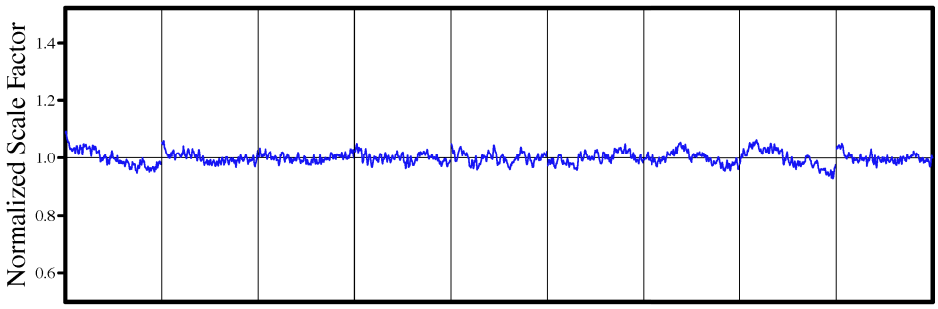
			neutron-diffraction structure of hydrogen cis-diacetyltetracarbonylrhenate, [cis-(OC)4Re(CH3CO)2]H: a metallaacetylacetone molecule". J. Am. Chem. Soc., 106:999-1003
11.	2.408	ABXNIH	Hussain M.S., Schlemper E.O. (1979). "A short and nearly symmetrical intramolecular hydrogen bond: x-ray and neutron diffraction analysis of 2,2-(1,3-diaminopropane)bis(2-methyl-3-butanone oximato)nickel(II) chloride hydrate". Inorg. Chem. 18:2275-2282
12.	2.410	MALAQZ03	Sequeira A., Rajagopal H., Gupta M.P., Vanhouteghem F., Lenstra A.T.H., Geise H.J. (1992). "Tetraaquabis(hydrogen maleato)zinc(II) by neutron diffraction and tetraaquabis(hydrogen maleato)nickel(II) by high-order X-ray diffraction". Acta Crystallogr. Sect.C 48:1192-1197
13.	2.412	FAVCEK01	Schlemper E.O., Murmann R.K., Hussain M.S. (1986). "Neutron and X-ray diffraction study of a macrocyclic nickel(II) complex with Ni^{II} in a pseudoaromatic chelate ring". Acta Crystallogr. Sect.C 42:1739-1743
14.	2.425 2.429 2.434 2.435 2.435	TACETA08 TACETA05 TACETA04 TACETA06 TACETA07	Piccoli P.M.B., Koetzle T.F., Schultz A.J., Zhurova E.A., Stare J., Pinkerton A.A., Eckert J., Hadzi D. (2008). "Variable Temperature Neutron Diffraction and X-Ray Charge Density Studies of Tetraacetylethane". J. Phys. Chem. A 112:6667-6677
15.	2.430	MALAQZ01	Antsyshkina A.S., Dikareva L.M., Porai-Koshits M.A., Fykin L.E., Dudarev V.Ya., Gusejnov M.G. (1982) Koord. Khim. (Russ.)(Coord.Chem.), 8, 1256
16.	2.435 2.442 2.448 2.464 2.468 2.475	DBEZLM07 DBEZLM10 DBEZLM08 DBEZLM09 DBEZLM12 DBEZLM11	Thomas L.H., Florence A.J., Wilson C.C. (2009). "Hydrogen atom behaviour imaged in a short intramolecular hydrogen bond using the combined approach of X-ray and neutron diffraction". New J. Chem. 33:2486-2490
17.	2.444 2.508	CIXKEA03	Kiyanagi R., Kojima A., Kimura H., Watanabe M., Noda Y., Mochida T., Sugawara T. (2005). "Phase Transition Scheme of Isolated Hydrogen-bonded Material h-MeHPLN Studied by Neutron and X-ray Diffraction". J. Phys. Soc. Jpn., 74:613-620
18.	2.445	NAHMAL01	Olovsson G., Olovsson I., Lehmann M.S. (1984). "Neutron diffraction study of sodium hydrogen maleate trihydrate, $NaH[C_4H_2O_4].3H_2O$, at 120 K". Acta Crystallogr. Sect.C 40:1521-1526
19.	2.450	TACETA01	Power L.F., Turner K.E., Moore F.H. (1975). "Refinement of the crystal and molecular structure of tetraacetylethane by neutron diffraction". J. Cryst. Mol. Struct. 5:59-66
20.	2.459	DBEZLM02	Jones R.D.G. (1976). "The crystal structure of the enol tautomer of 1,3-diphenyl-1,3-propanedione (dibenzoylmethane) by neutron diffraction". Acta Crystallogr. Sect.B 32:1807-1811
21.	2.472	AMBXPT10	Schlemper E.O., Fair C.K. (1977). "A short, highly asymmetrical intramolecular hydrogen bond: a neutron

			diffraction study of bis(2-amino-2-methyl-3-butanone oximato)platinum(II) chloride 3.5 hydrate $[Pt(C_5H_{11}N_2O)_2H]^+Cl^-.3.5H_2O$". Acta Crystallogr. Sect.B 33:2482-2489
22.	2.474 2.479 2.481 2.479 2.511 2.512	CODXAU04 RALBUB02	Ohhara T., Uekusa H., Ohashi Y., Tanaka I., Kumazawa S., Niimura N. (2001). "Direct observation of deuterium migration in crystalline-state reaction by single-crystal neutron diffraction. III. Photoracemization of 1-cyanoethyl cobaloxime complexes". Acta Crystallogr. Sect.B 57:551-559
23.	2.474	GUSRIV	Pleier A.-K., Herdtweck E., Mason S.A., Thiel W.R. (2003). "1-Aryl-3-(dimethylamino)propenones: Strong Proton Acceptors for Hydrogen Bonds". Eur. J. Org. Chem., 499-506
24.	2.479	AEBXNI10	Fair C.K., Schlemper E.O. (1978). "A neutron diffraction study of the short hydrogen bond in a tetradentate /a-amine oxime complex of nickel(II)". Acta Crystallogr. Sect.B 34:436-442
25.	2.485	BZOYAC01	Jones R.D.G. (1976). "The crystal and molecular structure of the enol form of 1-phenyl-1,3-butanedione (benzoylacetone) by neutron diffraction". Acta Crystallogr. Sect.B 32:2133-2136
26.	2.485 2.511	COMBUB02	Ohhara T., Ikeda S., Imura H., Uekusa H., Ohashi Y., Tanaka I., Niimura N. (2002). "Direct Observation of Deuterium Migration in Crystalline-State Reaction by Single Crystal Neutron Diffraction IV. "Hula-Twist" Rotation of a Long Alkyl Radical Produced by Photoirradiation". J. Am. Chem. Soc., 124: 14736-14740
27.	2.500	NMGCOB01	Englert U., Heger G., Kummerle E., Wang R. (1999). "Crystal structure of aquabis(dimethylglyoximato)nitrocobalt(III) based on neutron diffraction data". Z. Kristallogr., 214:71-74
28.	2.501	BZOYAC06	Herbstein F.H., Iversen B.B., Kapon M., Larsen F.K., Madsen G.K.H., Reisner G.M. (1999). "X-ray and neutron diffraction study of benzoylacetone in the temperature range 8-300 K: comparison with other *cis*-enol molecules". Acta Crystallogr. Sect.B 55:767-787
29.	2.502	BZOYAC04	Madsen G.K.H., Iversen B.B., Larsen F.K., Kapon M., Reisner G.M., Herbstein F.H. (1998). "Topological Analysis of the Charge Density in Short Intramolecular O−H···O Hydrogen Bonds. Very Low Temperature X-ray and Neutron Diffraction Study of Benzoylacetone". J. Am. Chem. Soc., 120:10040-10045
30.	2.532	DELHOS04	Hosoya T., Uekusa H., Ohashi Y., Ohhara T., Kuroki R. (2006). "A New Photoisomerization Process of the 4-Cyanobutyl Group in a Cobaloxime Complex Crystal Observed by Neutron Diffraction". Bull. Chem. Soc. Jpn. 79:692-701
31.	2.536	RACCYC03	Ohgo Y., Ohashi Y., Klooster W.T., Koetzle T.F. (1997). "Analysis of Hydrogen-Deuterium Exchange Reaction in a Crystal by Neutron Diffraction.". Enantiomer, 2:241-248
32.	2.550	HFULCA01	Fuess H., Lindner H.J. (1975). "Intramolekulare

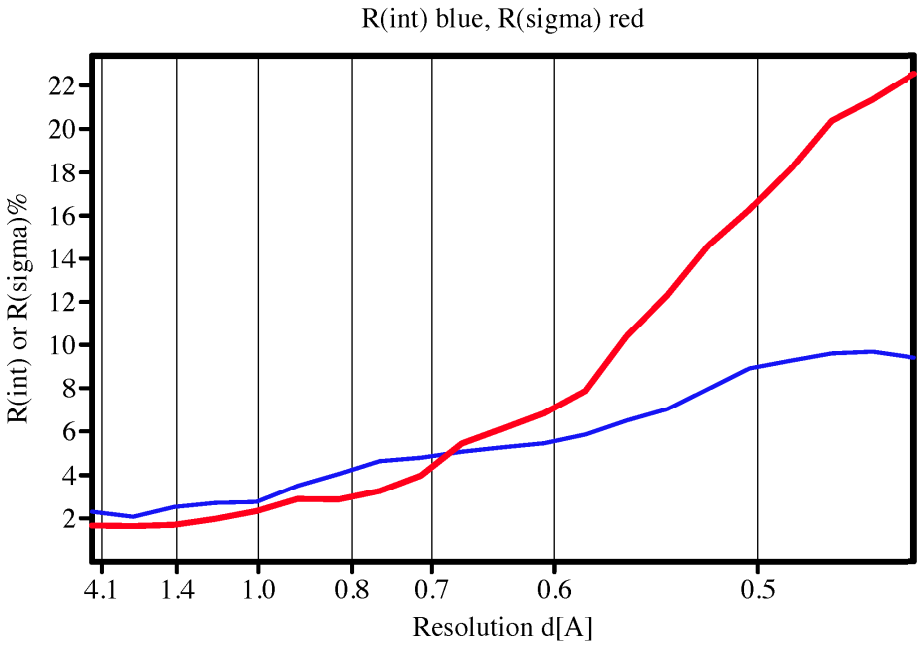
			Wasserstoffbrücke in 6-Hydroxy-1-fulvencarbaldehyd". Chem.Ber., 108:3096-3104
33.	2.558	GADBAP01	Hibbs D. E., Overgaard J., Piltz R.O. (2003). "X–N Charge density analysis of the hydrogen bonding motif in 1-(2-hydroxy-5-nitrophenyl)ethanone". Org. Biomol. Chem. 1:1191-1198
34.	2.570 2.583	DHNAPH17	Herbstein F.H., Kapon M., Reisner G.M., Lehman M.S., Kress R.B., Wilson R.B., Shiau W.-I., Duesler E.N., Paul I.C., Curtin D.Y. (1985). "Polymorphism of Naphthazarin and its Relation to Solid-State Proton Transfer. Neutron and X-Ray Diffraction Studies on Naphthazarin C". Proc. R. Soc. London, Ser.A, 399:295-319
35.	2.573 2.573 2.576	BESKAL11 BESKAL12 BESKAL10	Adam M.S., Gutmann M.J., Leech C.K., Middlemiss D.S., Parkin A., Thomas L.H., Wilson C.C. (2010). "Stability and cooperativity of hydrogen bonds in dihydroxybenzoic acids". New J. Chem., 34:85-91
36.	2.583	ZZZFQQ02	Cotton F.A., Falvello L.R., Murillo C.A., Schultz A.J. (1992). Eur. J. Solid State Inorg. Chem., 29:311
37.	2.598	PYRXCL01	Bacon G.E., Plant J.S. (1980). "Neutron structural refinement for pyridoxinium chloride, a component $C_8H_{12}ClNO_3$ of the vitamin B_6". Acta Crystallogr. Sect.B 36:1130-1136

c) Diagrams concerning the data quality of the X-ray diffraction experiment on **1**

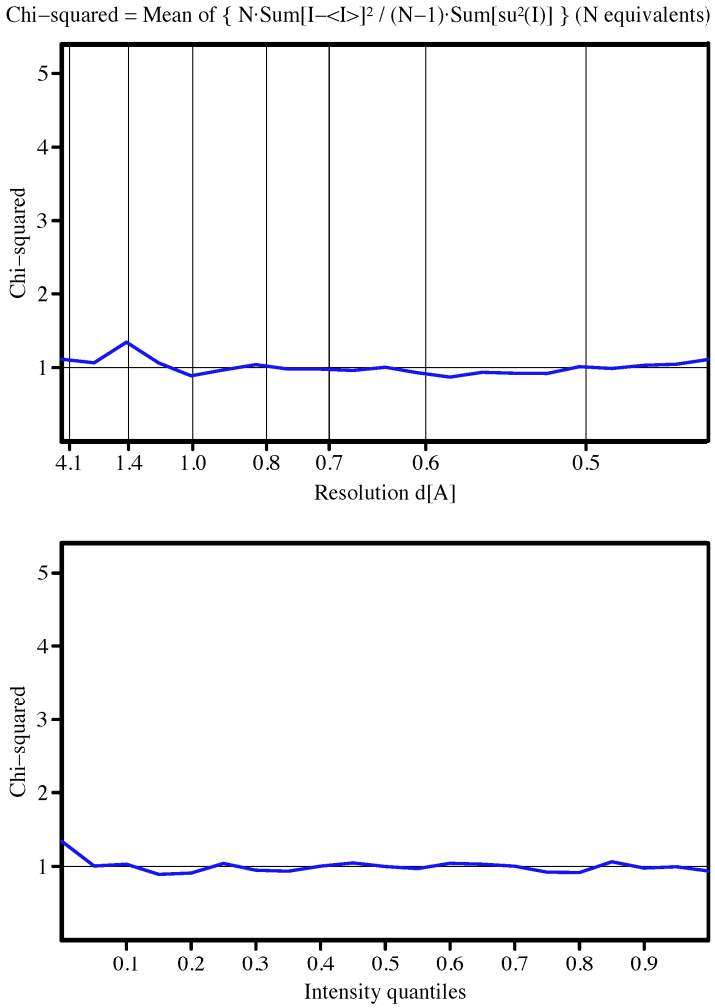
Normalized scale factor vs run number



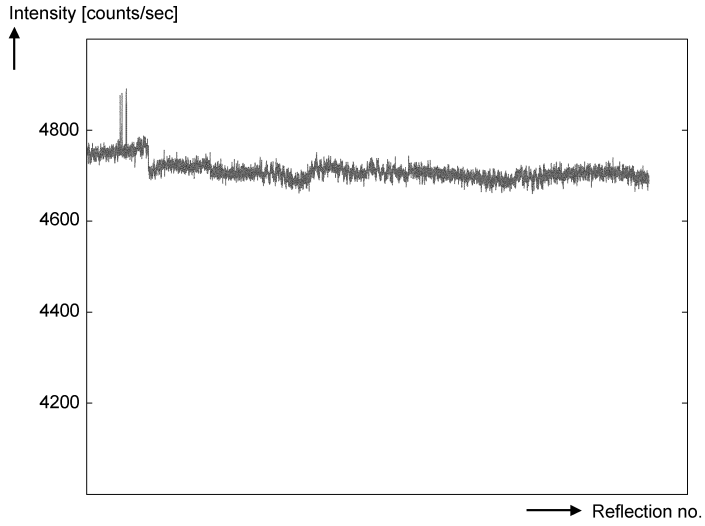
R(int) and R(sigma) as a function of the resolution



Diagrams concerning the data quality of the X-ray diffraction experiment on **1**



d) Beam stability in the neutron diffraction experiment on **1**



e) Table S1. Completeness of X-ray and neutron intensity data for **1**.

Space Group : P-1

Unit Cell (CIF) : 7.0689 8.5538 13.0736 90.426 105.118 102.425

SHELX WGHT Pars. : 0.0250 0.0000

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PLATON-(Version 310310) FCF-File Validation for: X-ray 1 Wavelength (Ang) : 0.71073

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Resolution & Completeness Statistics (Cumulative and Friedel Pairs Averaged)

==========

Theta	sin(th)/Lambda	Complete	Expected	Measured	Missing
20.82	0.500	0.999	1552	1551	1
23.01	0.550	1.000	2073	2072	1
25.24	0.600	1.000	2702	2701	1
27.51	0.650	0.999	3414	3412	2
29.84	0.700	0.997	4253	4241	12
32.21	0.750	0.994	5261	5229	32
34.65	0.800	0.990	6391	6328	63
37.17	0.850	0.986	7643	7534	109
39.77	0.900	0.978	9086	8890	196
42.47	0.950	0.968	10686	10348	338
45.29	1.000	0.956	12465	11914	551
48.27	1.050	0.943	14410	13587	823
51.43	1.100	0.928	16600	15408	1192
52.20	1.112	0.924	17100	15808	1292

==========

PLATON-(Version 310310) FCF-File Validation for: neutron 1 Wavelength (Ang) : 0.86800

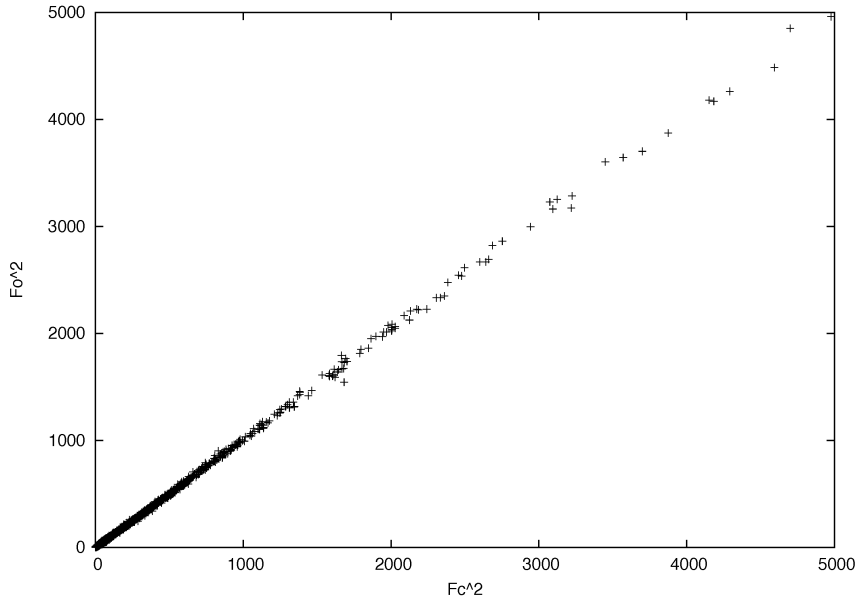
==========

Resolution & Completeness Statistics (Cumulative and Friedel Pairs Averaged)

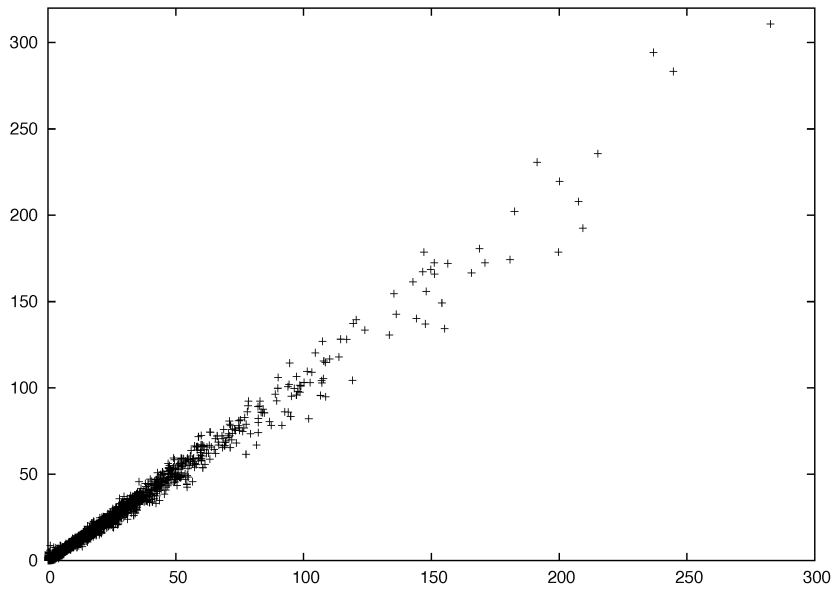
==========

Theta	sin(th)/Lambda	Complete	Expected	Measured	Missing
25.72	0.500	0.882	1552	1369	183
28.52	0.550	0.882	2073	1829	244
31.39	0.600	0.869	2702	2348	354
34.35	0.650	0.849	3414	2897	517
37.42	0.700	0.845	4253	3594	659
40.62	0.750	0.854	5237	4472	765
43.98	0.800	0.848	6274	5319	955
45.43	0.821	0.831	6690	5560	1130

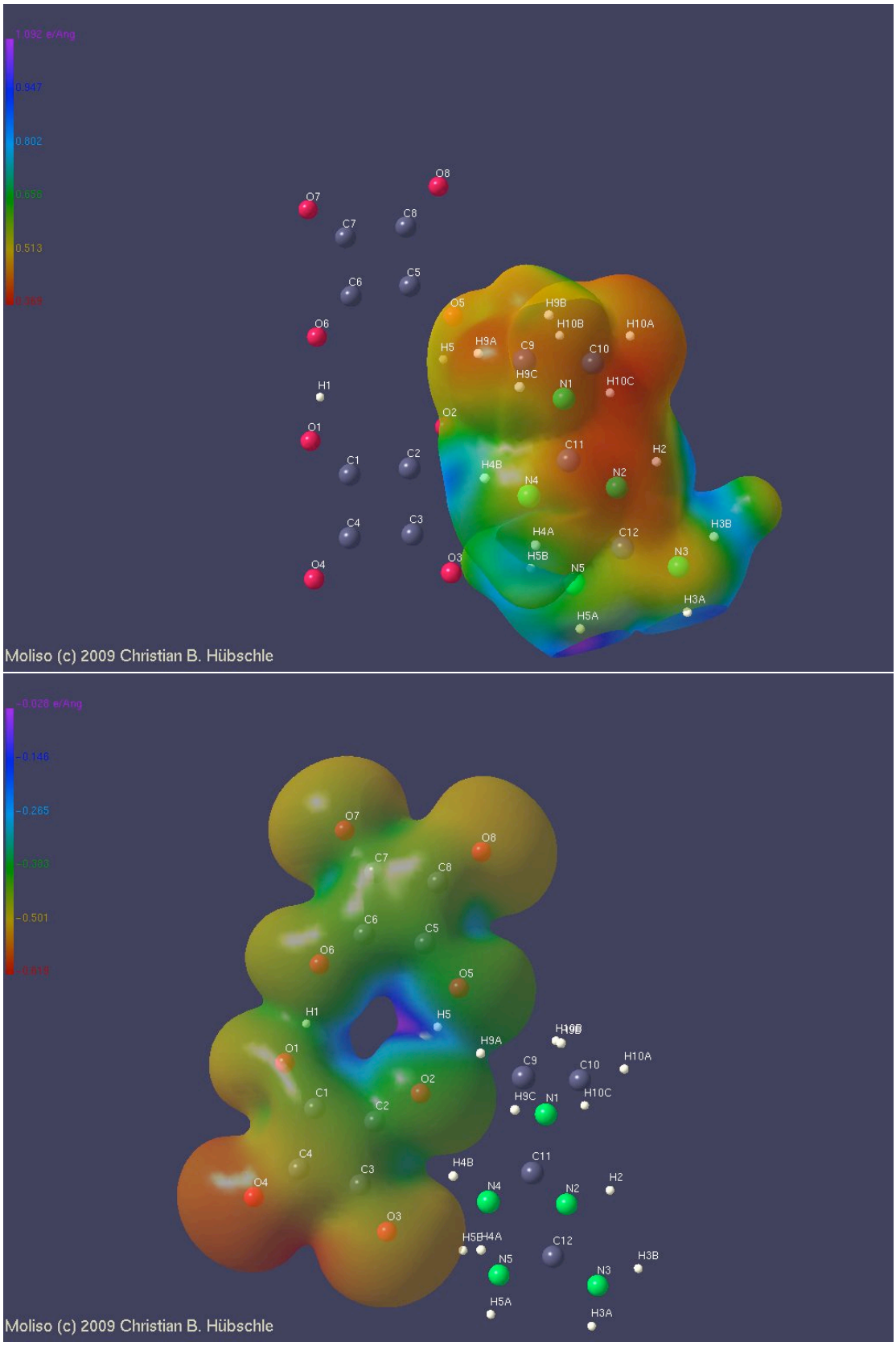
f) Scatterplot F^2_{obs} vs F^2_{calc} for X-ray refinement result on **1**.



g) Scatterplot F^2_{obs} vs F^2_{calc} for neutron refinement result on **1**.



h) Isosurface maps showing the electrostatic potentials of a dication (top) and two H-bonded monoanions (bottom) in the crystal structure of **1**. (MOLISO, C. B. Hübschle, P. Luger, J. Appl. Crystallogr. **2006**, 39, 901)



i) Table S2. Interatomic distances, bond paths R_{ij}, d_1, d_2 (Å), electron densities ρ (eÅ^{-3}) and Laplacian of the electron density $\nabla^2\rho$ (eÅ^{-5}) in the bond critical points for covalent bonds in **1**

Bond	Interat. dist (Å)	R_{ij}(Å)	d_1(Å)	d_2(Å)	ρ (eÅ^{-3})	$\nabla^2\rho$ (eÅ^{-5})
O(1)-C(1)	1.2925(6)	1.2927	0.8595	0.4332	2.47(2)	-20.7(2)
O(1)-H(1)	1.0480(5)	1.0489	0.8372	0.2117	1.78(2)	-26.9(2)
O(2)-C(2)	1.2431(6)	1.2437	0.8252	0.4185	2.85(2)	-25.0(2)
O(3)-C(3)	1.2307(6)	1.2307	0.8219	0.4088	2.78(2)	-11.86(8)
O(4)-C(4)	1.2464(6)	1.2464	0.8215	0.4249	2.87(2)	-26.65(8)
O(5)-C(5)	1.3019(6)	1.3023	0.8702	0.4321	2.39(2)	-18.71(7)
O(5)-H(5)	1.0210(5)	1.0216	0.8466	0.1750	1.89(2)	-45.0(2)
O(6)-C(6)	1.2476(6)	1.2483	0.8267	0.4216	2.77(2)	-22.45(8)
O(7)-C(7)	1.2351(6)	1.2353	0.8239	0.4114	2.85(2)	-18.65(8)
O(8)-C(8)	1.2334(6)	1.2334	0.8252	0.4083	2.87(2)	-14.25(8)
N(1)-C(9)	1.4613(6)	1.4616	0.9027	0.5589	1.81(2)	-14.38(5)
N(1)-C(10)	1.4592(6)	1.4594	0.8754	0.5840	1.83(2)	-14.05(4)
N(1)-C(11)	1.3177(6)	1.3177	0.8836	0.4341	2.52(2)	-20.4(8)
N(2)-C(11)	1.3779(6)	1.3787	0.8901	0.4886	2.19(2)	-24.93(6)
N(2)-C(12)	1.3768(6)	1.3775	0.8460	0.5315	2.39(2)	-31.88(5)
N(2)-H(2)	1.0340(5)	1.0340	0.8192	0.2148	2.02(2)	-28.9(2)
N(3)-C(12)	1.3227(7)	1.3228	0.8864	0.4364	2.52(2)	-23.36(8)
N(3)-H(3A)	1.0140(5)	1.0148	0.7893	0.2255	2.14(2)	-37.8(2)
N(3)-H(3B)	1.0170(5)	1.0171	0.7883	0.2289	2.19(2)	-37.5(2)
N(4)-C(11)	1.3293(6)	1.3294	0.8872	0.4422	2.47(2)	-24.17(7)
N(4)-H(4A)	1.0150(5)	1.0159	0.8005	0.2154	2.15(2)	-37.5(2)
N(4)-H(4B)	1.0110(5)	1.0112	0.7879	0.2232	2.18(2)	-35.6(2)
N(5)-C(12)	1.3118(6)	1.3119	0.8826	0.4294	2.58(2)	-19.71(8)
N(5)-H(5A)	1.0230(5)	1.0232	0.8050	0.2181	2.16(2)	-39.4(2)
N(5)-H(5B)	1.0200(5)	1.0209	0.8069	0.2140	1.99(2)	-34.2(2)
C(1)-C(2)	1.4480(6)	1.4528	0.7358	0.7170	2.035(9)	-17.91(3)
C(1)-C(4)	1.4376(6)	1.4415	0.7612	0.6802	2.108(10)	-19.35(3)
C(2)-C(3)	1.4905(5)	1.4933	0.7347	0.7587	1.913(9)	-16.31(3)

C(3)-C(4)	1.4785(6)	1.4808	0.7300	0.7507	1.904(9)	-14.58(3)
C(5)-C(6)	1.4322(6)	1.4362	0.7294	0.7068	2.060(10)	-18.15(3)
C(5)-C(8)	1.4534(6)	1.4567	0.7503	0.7064	2.047(10)	-19.08(3)
C(6)-C(7)	1.4779(6)	1.4807	0.7473	0.7334	1.933(9)	-16.17(3)
C(7)-C(8)	1.4964(6)	1.4999	0.7400	0.7599	1.861(9)	-14.97(3)
C(9)-H(9A)	1.0690(5)	1.0692	0.7537	0.3156	1.90(2)	-21.94(6)
C(9)-H(9B)	1.0870(5)	1.0871	0.7261	0.3609	1.83(2)	-19.73(5)
C(9)-H(9C)	1.0810(5)	1.0815	0.7501	0.3314	1.84(2)	-19.26(5)
C(10)-H(10A)	1.0760(6)	1.0760	0.7573	0.3187	1.84(2)	-19.59(5)
C(10)-H(10B)	1.0760(5)	1.0760	0.7070	0.3691	1.86(2)	-19.58(4)
C(10)-H(10C)	1.0880(5)	1.0881	0.7599	0.3282	1.84(2)	-19.03(6)

j) Table S3. Interatomic distances, bond paths R_{ij}(Å), electron densities ρ (eÅ^{-3}), Laplacian of the electron density $\nabla^2\rho$ (eÅ^{-5}), kinetic (*G, au*) and potential energy density *(V, au*) in the bond critical points for hydrogen bonds in **1**

	$d_{D\cdots A}$ (Å)	$d_{D\text{-}H}$ (Å)	$d_{H\cdots A}$ (Å)	< D- H···A°	ρ_{bcp} (e/Å^3) (H···A)	$\nabla^2\rho$ (*e*/Å^5)	*G(a.u.)*	*V(a.u.)*	*G/ρ(a.u.)*
O1–H1···O6	2.447(3)	1.048(4)	1.421(4)	164.6(4)	0.64(2)	2.43(5)	0.073	-0.121	0.770
O5–H5···O2	2.568(3)	1.022(4)	1.569(4)	164.5(4)	0.37(2)	3.14(4)	0.044	-0.055	0.803
N2–H2···O4[i]	2.742(2)	1.034(4)	1.779(4)	153.2(3)	0.24(2)	3.69(3)	0.038	-0.038	1.07
N5–H5b···O3	2.743(2)	1.019(4)	1.788(4)	154.5(3)	0.23(2)	2.78(3)	0.029	-0.029	0.851
N3–H3A···O8[iv]	2.824(2)	1.014(4)	1.849(4)	160.1(3)	0.23(2)	2.65(2)	0.028	-0.029	0.821
N3–H3B···O4[i]	2.789(2)	1.017(4)	1.893(4)	145.2(3)	0.18(2)	2.59(3)	0.025	-0.023	0.937
N5–H5A···O7[iv]	2.822(2)	1.023(4)	1.838(4)	160.5(4)	0.18(2)	2.87(3)	0.027	-0.024	1.01
N4–H4A···O3[ii]	2.856(2)	1.015(4)	1.900(4)	155.7(4)	0.17(2)	2.34(2)	0.023	-0.022	0.913
N4–H4B···O7[iii]	3.008(2)	1.010(4)	2.068(5)	154.1(4)	0.090(9)	2.21(2)	0.017	-0.012	1.27
C9–H9A···O6[iii]	3.211(3)	1.070(7)	2.265(7)	146.3(5)	0.072(4)	1.391(2)	0.011	-0.008	1.03
C10–H10B···O5	3.484(3)	1.076(6)	2.531(6)	147.2(4)	0.027(3)	0.749(2)	0.0055	-0.003	1.37

Symmetry operators: *i* = *x*-1, *y*-1, *z*; *ii* = 2-*x*, 1-*y*, 2-*z*; *iii* = 2-*x*, 1-*y*, 1-*z*; *iv* = *x*, *y*, 1+*z*