3,7-Diazabicyclo[3.3.1]nonane-2,6-diones: building of homo- and heterochiral crystals

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A strategy in the synthesis of 1,5- or 3,7-unsubstituted derivatives of the title dilactams has been developed, and the parent 3,7-diazabicyclo[3.3.1]nonane-2,6-dione **4** was synthesised; the hydrogen bonding generated self-assembly as a heterochiral infinite diagonal zigzag tape was found in the crystal of **2** (space group $P2_1/c$) whereas a homochiral helical architecture was observed in **4** (space group $P2_12_12_1$).

In order to design molecules which are capable of the homochiral self-assembly to form crystalline conglomerates, bicyclic dilactams of the 3,7-diazabicylo[3.3.1]nonane-2,6-dione series were examined. It is anticipated *a priori* that the hydrogen-bonded self-assembly will occur either in helical suprastructures, as in the case of chiral glycouril,[1] or in heterochiral diagonal zigzag tapes, as in dilactam **A** of the bicyclo[3.3.0]-octane series[2] (*cf.* ref. 3). Both of these opportunities can be realised.

The only examples reported earlier are derivatives of 3,7-dialkyl-3,7-diazabicyclo[3.3.1]nonane-2,6-dione-1,5-dicarboxylic acids.[4,5] To synthesise the target compounds, we have used a recently developed strategy for preparing similar dilactams **A** and **B** of the 3,7-diazabicyclo[3.3.0]octane-2,6-dione series[2] (Scheme 1).

Dilactam **1** was synthesised on the basis of tetraethyl propane-1,1,3,3-tetracarboxylate[6] using the method proposed by Knowles and co-authors[4] with the new aminomethylating reagent 1,3,5-tris(4-methoxybenzyl)hexahydro-1,3,5-triazine.[2] N-Deprotection of **1** with cerium ammonium nitrate (CAN)[2] gives dilactam diester, and its subsequent deesterification leads to dilactam diacid **3**. The latter was thermally decarboxylated to form parent dilactam **4**. The opposite sequence of transformations has also been performed; it includes deesterification of **1** into 3,7-disubstituted dilactam diacid **5** and its decarboxylation into 3,7-disubstituted dilactam **6**. The latter was a matter of particular interest because its analogue of the bicyclo[2.2.2]-octane series forms a conglomerate.[7] However, product **6** was not found to be crystalline.

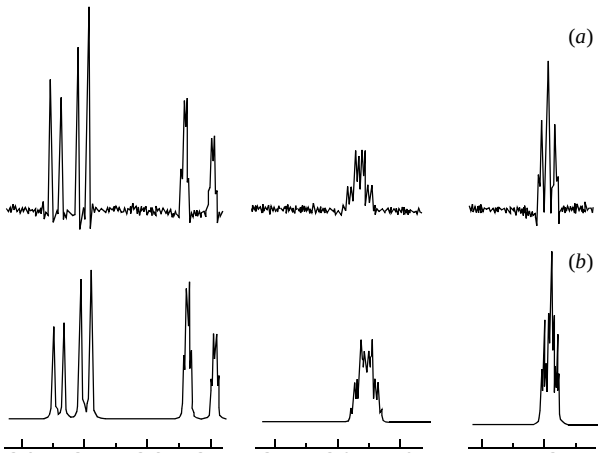


Figure 1 ^{1}H NMR spectra of **4**: (*a*) an experimental spectrum in CD_3OD and (*b*) a spectrum calculated by CALM.

All of the compounds were characterised by spectroscopic data† (Figure 1). The structures of **2** and **4** were confirmed by X-ray diffraction analysis‡ (Figures 2 and 3).

Experimental values of the vicinal spin–spin coupling constants for **4** (*cf.* the data for bicyclo[3.3.1]nonane-2,6-diones[8]) are in agreement with those calculated by the general Karplus equation

† *Characteristics and spectroscopic data*. NMR spectra were recorded on a Bruker WM-400 spectrometer [at 400.13 (^{1}H) and 100.62 MHz (^{13}C)], ^{1}H NMR spectrum of **4** was recorded on a Bruker AM-300 spectrometer (at 300.13 MHz) and calculated by the CALM program (Resonance, Moscow, 1993).

1: yield 55%, mp 131–132 °C (Et_2O–petroleum ether). ^{1}H NMR ($CDCl_3$) δ: 1.24 (t, 6H, 2Me, ^3J 7.5 Hz), 2.69 (t, 2H, 9-CH_2, $^4J_{obs}$ 1.2 Hz), 3.63 (dt, 2H, 4,8-CH_b, ^2J –12.8 Hz, $^4J_{obs}$ 1.2 Hz), 3.77 (s, 6H, 2MeO), 3.80 (d, 2H, 4,8-CH_a, ^2J –12.8 Hz), 4.20 (m, 4H, 2CH_2O, ABX_3 spectrum), 4.52 (m, 4H, 3,7-NCH_2, AB spectrum, $\Delta\nu$ 26.0, ^2J –14.6 Hz), 6.86 and 7.13 (d and d, 8H, 2C_6H_4, ^3J 9.0 Hz). ^{13}C NMR ($CDCl_3$) δ: 13.72 (qt, Me, ^1J 127.2 Hz, ^2J 2.9 Hz), 33.31 (br. t, 9-CH_2, ^1J 136.6 Hz), 49.73 (s, 1,5-C), 49.90 (t, 3,7-NCH_2, ^1J 138.8 Hz), 52.63 (br. tt, 4,8-CH_2, ^1J 145.3 Hz, ^3J 4.4 Hz), 54.96 (q, MeO, ^1J 143.8 Hz), 68.80 (tq, CH_2O, ^1J 148.2 Hz, ^2J 4.4 Hz), 113.9 (dd, 3′-C, ^1J 159.0 Hz, ^2J 4.4 Hz), 127.6 (m, 1′-C), 129.05 (dm, 2′-C, ^1J 157.6 Hz), 158.95 (br. s, 4′-C), 166.2 (br. s, CO_2Et), 168.6 (br. s, 2,6-CO).

2: yield 49%, mp 203–205 °C (EtOH). ^{1}H NMR ($CDCl_3$) δ: 1.29 (t, 6H, 2Me, ^3J 7.0 Hz), 2.70 (t, 2H, 9-CH_2, $^4J_{obs}$ 1.2 Hz), 3.73 (ddt, 2H, 4,8-H_b, ^2J –12.5 Hz, $^3J_{Hbe}$ 4.3 Hz, $^4J_{bd'(d)obs}$ 1.2 Hz), 3.84 (d, 2H, 4,8-H_a, $^2J_{ab}$ –12.5 Hz), 4.25 (q, 4H, 2CH_2O, ^3J 7.0 Hz). ^{13}C NMR (CD_3OD) δ: 13.90 (qt, Me, ^1J 127.2 Hz, ^2J 2.9 Hz), 32.50 (tt, 9-CH_2, ^1J 137.0 Hz, ^3J 7.0 Hz), 47.35 (tt, 4,8-CH_2, ^1J 147.0 Hz, ^3J 5.2 Hz), 50.10 (s, 1,5-C), 62.75 (tq, CH_2O, ^1J 148.2 Hz, ^2J 4.4 Hz), 170.0 and 170.2 (s and s, 2,6-CO and CO_2).

3: yield 86%, mp 250–252 °C (decomp., from H_2O). ^{1}H NMR (CD_3OD) δ: 2.70 (s, 2H, 9-CH_2), 3.65 (m, 4H, 4,8-CH_2, AB spectrum, $\Delta\nu$ 68.0, ^2J –13.0 Hz).

4: yield 67.4%, mp 350 °C (MeOH). ^{1}H NMR (CD_3OD) δ: 2.30 (m, 2H, dd′, ^2J –13.0 Hz, $^3J_{dc'} = {}^3J_{d'c} = 3.72$ Hz, $^3J_{dc} = {}^3J_{d'c'} = 2.57$ Hz, $^4J_{db'} = {}^4J_{d'b} = 2.02$ Hz, $^4J_{da'} = {}^4J_{d'a} = -0.33$ Hz), 2.97 (m, 2H, cc′, $^3J_{ca} = {}^3J_{c'a'} = 5.02$ Hz, $^3J_{cb} = {}^3J_{c'b'} = 1.45$ Hz, $^4J_{cc'}$ –1.50 Hz), 3.52 (m, 2H, bb′, $^2J_{ba} = {}^2J_{b'a'} = -13.04$ Hz), 3.72 (m, 2H, aa′). ^{13}C NMR (D_2O) δ: 22.0 (t, 9-CH_2), 33.0 (d, 1,5-CH), 43.9 (t, 4,8-CH_2), 173.6 (s, 2,6-CO).

5: yield 73%, mp 220 °C. ^{1}H NMR (CD_3OD) δ: 2.70 (s, 2H, 9-CH_2), 3.70 (m, 4H, 4,8-CH_2, AB spectrum, $\Delta\nu$ 60.0, ^2J –12.7 Hz), 3.78 (s, 6H, 2MeO), 4.50 (m, 4H, 3,7-NCH_2, AB spectrum, $\Delta\nu$ 76.0, ^2J –14.3 Hz), 6.87 and 7.12 (d and d, 8H, 2C_6H_4, ^3J 8.5 Hz), 7.9 (s, 2H, 2CO_2H).

6: yield 16%, viscous oil. ^{1}H NMR ($CDCl_3$) δ: 2.09 (t, 2H, 9-CH_2, ^3J 3.0 Hz), 2.91 (dtd, 2H, 1,5-CH, $^3J_{ac}$ 4.7 Hz, $^3J_{cd}$ 3.0 Hz, $^3J_{bc}$ 1.0 Hz), 3.40 (dd, 2H, 4,8-$H_aH_{a'}$, $^2J_{ab}$ –12.1 Hz, $^3J_{ac}$ 4.7 Hz), 3.48 (dd, 2H, 4,8-$H_bH_{b'}$, $^2J_{ab}$ –12.1 Hz, $^3J_{bc}$ 1.0 Hz), 3.80 (s, 6H, 2MeO), 4.50 (m, 4H, 3,7-NCH_2, AB spectrum, $\Delta\nu$ 120.0, ^2J –14.1 Hz), 6.82 and 7.11 (d and d, 8H, 2C_6H_4, ^3J 8.7 Hz). ^{13}C NMR ($CDCl_3$) δ: 25.44 (t, 9-C, ^1J 133.7 Hz), 36.90 (d, 1,5-C, ^1J 137.5 Hz), 49.10 (tt, 3,7-NCH_2, ^1J 138.6 Hz, ^3J 4.0 Hz), 51.10 (t, 4,8-CH_2, ^1J 141.3 Hz), 55.10 (q, MeO, ^1J 143.7 Hz), 113.9 (dd, 3′-C, ^1J 160.2 Hz, ^2J 5.0 Hz), 128.16 (s, 1′-C), 129.0 (ddt, 2′-C, ^1J 158.0 Hz, ^2J 7.0 Hz, ^3J 3.5 Hz), 158.9 (s, 4′-C), 169.8 (s, 2,6-CO).

Table 1 Vicinal spin–spin coupling constants and dihedral angles in the investigated molecules.

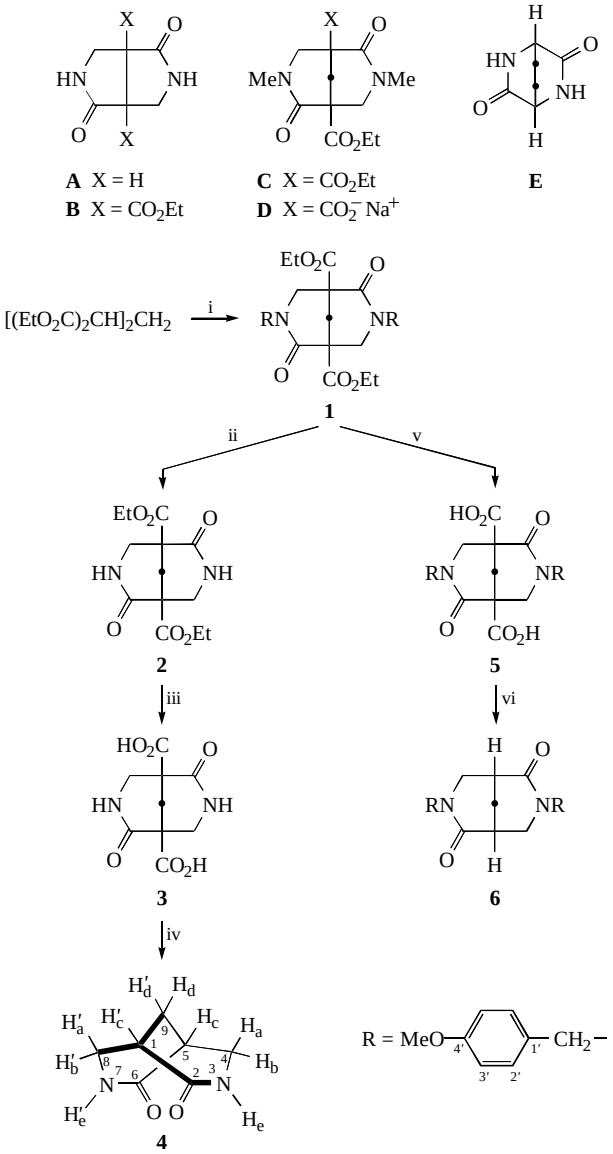
Protons and carbons	^3J/Hz for **4** (and **2**, **6**, **C**)		Dihedral angles (°)	
	NMR	Calculated[a]	X-Ray for **4** (and **2**)	MM2 for **4**
ac	5.02 (4.7)[b]	3.93	51.7	50.0
bc	1.45 (1.0)[b]	1.79	68.2	68.0
cd	2.57 (3.0)[b]	2.06	64.8	67.0
cd′	3.72	4.23	50.3	51.0
ae	(0.0)[c] (0.0)[d]	~1.0	76.0 (71.3)	72.8
be	(4.3)[c] (2.2)[d]	~4.0	42.2 (48.4)	42.7
C(9)a	(0.0)[c] (0.0)[e]	1.2	70.1 (71.0)	69.0
C(9)b	(7.0)[c] (7.0)[e]	9.3	171.1 (171.1)	172.7
C(4)d′ [C(8)d]	(5.2)[c] (4.4)[e]	9.2	172.6 (170.6)	171.0
C(4)d [C(8)d′]		4.1	52.5 (52.7)	51.9

[a]By the general Karplus equation from the MM2 geometry; $^3J_{HH}$, by the general Karplus equation,[9] $^3J_{CH}$ by the Karplus curve for norbornane.[10] [b]For **6**. [c]For **2**. [d] $^3J_{CH}$ for carbon of MeN with a and b protons in compound **C**.[5] [e]For **C**.[5]

($^3J_{HH}$)[9] and by an equation for norbornanes ($^3J_{CH}$)[10] from the MM2 geometry, which is similar to the experimental one (Table 1). Similar values of the dihedral angles HH and HC and the corresponding spin–spin coupling constants ^3J were found for **2**, **4** and other examined compounds. The virtual spin–spin coupling constants of the carbons C(4) and C(8) with protons at C(9) are approximately equal to the half-sum of the constants $^3J_{4Cd'}$ and $^3J_{4Cd}$. The long-range virtual spin–spin coupling constant $^4J_{obs}$ = 1.2 Hz, of protons H_b, H'_b at C(4) and C(8) with protons H_d, H'_d at C(9) is observed for **1**, **2**, **4** and **C** (at {MeN})[5] (*cf.* ref. 8). It was shown earlier[5] that desymmetrisation of the system (for example, in going from **C** to **D**) resulted in the disappearance of virtual coupling, and the real coupling constants ($^4J_{HH}$ = 2.2 Hz) for each link appeared; they are about two times as large and correspond to those for propane calculated using the dihedral angles C(9)–b and C(4)–d′ close to 180° (Table 1).[11]

The bond lengths and bond angles in the crystal structures of **2** and **4** are very similar to those in the previously investigated derivatives **A** and **B** of dilactams of the bicyclo[3.3.0]octane series.[2] The angles between five-membered rings in the structures of **2** and **4** are 72.3° and 71.8°, respectively, and are significantly smaller in comparison with the corresponding values in the **A** and **B** structures (110.3°).[2] Both structures **2** and **4** are twisted; the pseudotorsion angles C(3)–C(4)–C(1)–C(2) and C(5)–C(4)–C(1)–C(6) are equal to 14.6°, 13.8° and 15.8°, 14.9°, respectively.

‡ *Crystallographic data for* **2** *and* **4** at 25 °C: crystals of $C_{13}H_{18}N_2O_6$ **2** are monoclinic, space group $P2_1/c$, a = 11.119(2) Å, b = 11.013(2) Å, c = 11.974(3) Å, β = 98.82(2)°, V = 1448.9(5) Å^3, Z = 4, M = 298.29, d_{calc} = 1.367 g cm^{-3}, μ(MoKα) = 1.09 cm^{-1}, F(000) = 632; crystals of $C_7H_{10}N_2O_2$ **4** are orthorhombic, space group $P2_12_12_1$, a = 6.603(4) Å, b = 9.841(4) Å, c = 10.704(3) Å, V = 695.5(5) Å^3, Z = 4, M = 154.17, d_{calc} = 1.472 g cm^{-3}, μ(MoKα) = 1.10 cm^{-1}, F(000) = 328. Intensities of 3525 reflections for **2** and 951 reflections for **4** were measured on a Siemens P3 diffractometer at 25 °C (λ MoKα radiation, $\theta/2\theta$-scan technique, $2\theta_{max}$ 56°) and 3698 (for **2**) or 951 (for **4**) independent reflections were used in further calculations and refinement. The structures were solved by the direct method and refined by a full-matrix least-squares against F^2 in the anisotropic–isotropic approximation. Hydrogen atoms were located from the difference Fourier synthesis and refined in the isotropic approximation. The refinement converged to wR_2 = 0.1081 and COF = 0.885 for all independent reflections [R_1 = 0.0385 is calculated against F for the 2212 observed reflections with $I > 2\sigma(I)$] for the structure of **2** and to wR_2 = 0.0783 and COF = 1.077 for all independent reflections [R_1 = 0.0294 is calculated against F for the 907 observed reflections with $I > 2\sigma$(I)] for the structure of **4**. All calculations were performed on an IBM PC/AT using the SHELXTL PLUS 5.0 program. Atomic coordinates, thermal parameters, bond lengths and bond angles have been deposited at the Cambridge Crystallographic Data Centre (CCDC). For details, see 'Notice to Authors', *Mendeleev Commun.*, 1999, Issue 1. Any request to the CCDC for data should quote the full literature citation and the reference number 1135/44.



Scheme 1 *Reagents and conditions*: i, 1,3,5-tris(4-methoxybenzyl)hexahydro-1,3,5-triazine[2] and CF_3CO_2H, 24 h at 100–120 °C; ii, $Ce(NH_4)_2(NO_3)_6$ (CAN) in MeCN–H_2O, then $NaHCO_3$; iii, KOH in MeOH, then HCl; iv, 20 min at 240–270 °C and 25 min at 290–300 °C *in vacuo* (20 Torr); v, KOH in EtOH, 12 h at 20 °C, separation of the precipitated K salt of **5** and its treatment with CF_3CO_2H in H_2O; vi, 20 min at 240–270 °C and 25 min at 290–300 °C *in vacuo* (20 Torr) with sublimation of **6**.

It was found that molecules of **2** are capable of self-assembling as a heterochiral H-bonded infinite diagonal zigzag tape[2] directed along the crystallographic *c* axis (Figure 4). The H-bond characteristics are N(1)–H(1N)···O(2′), (x, 1/2 – y, 1/2 + z); N(1)···O(2′), 2.886(3) Å; N(1)–H(1N)–O(2″), 167°; N(2)–H(2N)···O(1′), (x, 1/2 – y, –1/2 + z); N(2)···O(1′), 2.868(3) Å; N(2)–H(2N)–O(1′), 171°.

Note that as a result of the packing effects ethyl groups in the CO_2Et moiety are characterised by different conformations with the torsion angles C(10)–C(9)–O(4)–C(8) and C(13)–C(12)–O(6)–C(11) equal to 87.4° and 175.6°, respectively. Taking into account that the steric effect of the bulky CO_2Et groups can play a significant role in the formation of the crystal structure (as it was observed early in **A** and **B**), we expected the desirable helical-type structure in parent dilactam **4**. Indeed, as we found by X-ray diffraction analysis, dilactam **4** forms a crystalline conglomerate.‡ Similarly to the crystal structure of **A** and in contrast to that of **2**, each molecule in the structure of **4** forms H-bonds with four neighbouring molecules. As a result, the molecules of dilactam **4** are assembled by two H-bonded orthogonal helices [N(1′)–H(1N′)···O(1), (1/2 +x, 3/2 – y, 1 – z); N(1′)···O(1), 2.863(3) Å; N(1′)–H(1N′)–O(1), 172 Å; N(2)–

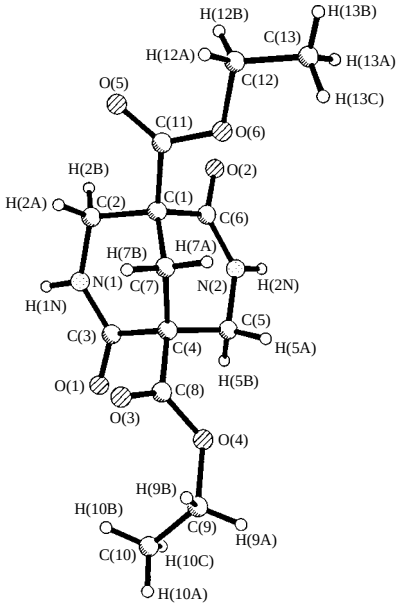


Figure 2 Molecular structure of **2**. Selected bond lengths (Å): O(1)–C(3) 1.234(2), O(2)–C(6) 1.236(2), N(1)–C(2) 1.457(2), N(1)–C(3) 1.336(2), N(2)–C(6) 1.335(2), N(2)–C(5) 1.459(2); selected bond angles (°): C(3)–N(1)–C(2) 127.2(1), C(6)–N(2)–C(5) 128.2(1), C(7)–C(1)–C(6) 112.0(1), C(7)–C(1)–C(2) 108.3(1), C(6)–C(1)–C(2) 109.0(1), N(1)–C(2)–C(1) 111.2(1), O(1)–C(3)–N(1) 122.4(1), O(1)–C(3)–C(4) 119.3(1), N(1)–C(3)–C(4) 118.2(1), C(7)–C(4)–C(5) 108.9(1), C(7)–C(4)–C(3) 112.2(1), C(5)–C(4)–C(3) 108.2(1), N(2)–C(5)–C(4) 110.6(1), O(2)–C(6)–N(2) 122.9(1), O(2)–C(6)–C(1) 119.2(1), N(2)–C(6)–C(1) 117.9(1), C(4)–C(7)–C(1) 107.3(1).

H(2N)···O(2), (1/2 + *x*, 3/2 – *y*, 2 – *z*); N(2)···O(2), 2.879(2) Å; N(2)–H(2N)–O(2′), 158°] into homochiral 'corrugated' layers parallel to the crystallographic *ac* plane (Figure 5). The crystal packing observed in **4** is very similar to the crystal structure of (–)-**E**.[7] Thus, starting from an incorrect assumption (the possibility of formation of a helical type structure[1]), as well as Lehn and co-workers (who expected the formation of a six-membered ring architecture in **E**[7]), we, nevertheless, have achieved our main goal to obtain the desirable conglomerate crystals of **4**. It should be noted that conglomerates were also found in the cases of 3,3,7,7-tetrabromobicyclo[3.3.1]nonane-2,6-dione (space group $P2_12_12_1$)[8] and 4-hydroxy-5-ethyl-6-carbamoyl-7-amino-3-azabicyclo[3.2.1]oct-6-en-2-one dihydrate (space group *P*1).[12]

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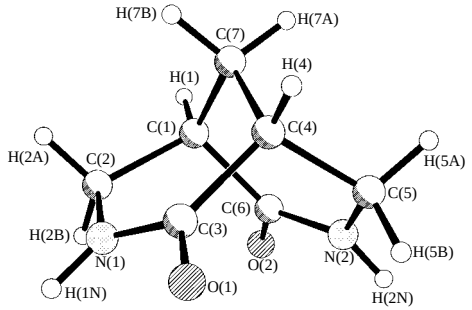

Figure 3 Molecular structure of **4**. Selected bond lengths (Å): O(1)–C(3) 1.231(2), O(2)–C(6) 1.236(2), N(1)–C(3) 1.335(2), N(1)–C(2) 1.454(2), N(2)–C(6) 1.338(2), N(2)–C(5) 1.460(2); selected bond angles (°): C(3)–N(1)–C(2) 126.3(1), C(6)–N(2)–C(5) 126.9(1), C(7)–C(1)–C(6) 112.4(1), C(7)–C(1)–C(2) 107.9(1), C(6)–C(1)–C(2) 109.5(1), N(1)–C(2)–C(1) 111.4(1), O(1)–C(3)–N(1) 122.1(2), O(1)–C(3)–C(4) 119.7(2), N(1)–C(3)–C(4) 118.2(1), C(3)–C(4)–C(7) 113.5(1), C(3)–C(4)–C(5) 108.6(1), C(7)–C(4)–C(5) 108.1(1), N(2)–C(5)–C(4) 110.5(1), O(2)–C(6)–N(2) 121.9(1), O(2)–C(6)–C(1) 120.2(1), N(2)–C(6)–C(1) 117.9(1), C(4)–C(7)–C(1) 106.8(1).

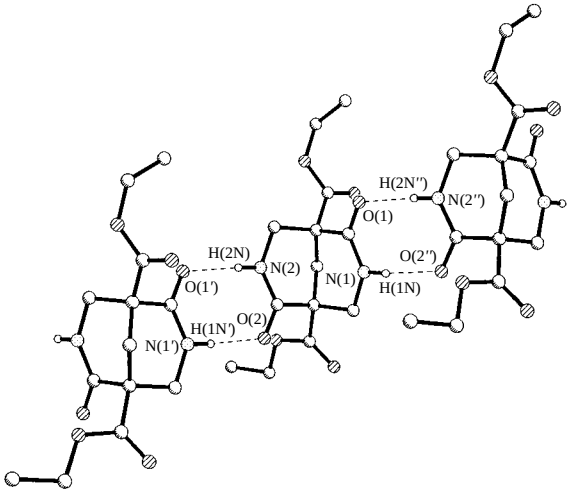

Figure 4 Diagonal zigzag tape in the crystal structure of **2**.

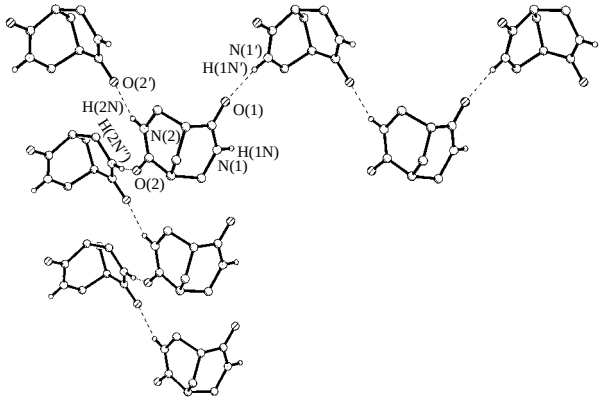

Figure 5 Schematic diagram of the homochiral layer formation in the crystal structure of **4**. For clarity, only molecules belonging to two orthogonal helices were selected.

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