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Supramolecular architectures constructed through self-assembly of a chalcone and substituted diazo-β-diketones

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Abstract

Organic compounds namely pyridyl chalcone viz. 3-[4-(3-oxo-3-pyridin-2-yl-propenyl)-phenyl]-1-pyridin-2-yl-propenone (L^1), *p*-cholorophenyldiazopentane-2,4-dione (L^2) and *p*-methyl phenyldiazopentane-2,4-dione (L^3) have been characterized by their single-crystal X-ray crystallographic studies. Several structural motifs resulting upon their self-association through probable non-covalent interactions have been discussed. The studies of related motifs found in Cambridge Structural Database are performed and the results are related to the structural data obtained for crystal structures reported here in.

Keywords: Crystal structure; Hydrogen bonding; Self-association

1. Introduction

Porous structures or open frameworks including removable solvents or exchangeable ions that provide new sizes, shapes and chemical environments have been of great interest in recent years, due to their potential applications [1,2] like mimicking zeolites etc. In recent years, efforts have been devoted to the synthesis and characterization of supramolecular complexes displaying a large variety of topologies [3]. Such structures can find diverse applications in the field of catalysis, host–guest chemistry, electrical and magnetic materials [4]. In this context, particular attention has been focused on metal assisted self-assembly of helical and box like metallic complexes [5]. Structural studies of complexes specially derived from Schiff base ligands have shown that CH-π and π–π interactions lead to the di or multi-helical topology of the complexes. Such structures originate from weak intermolecular (inter-strand) face to face and edge to face π–π interactions between aromatic rings [6]. Additionally, supramolecular approach has also been used to control reactivity in the organic solid that employs molecules in the form of linear templates to direct the [2+2] photo dimerization through network of hydrogen bonds [7]. Among various molecular interactions, the H-bond has received special attention due to its significance from biological view-point. Special importance is the creation of higher order structures of peptides and nucleic acids and the biochemical processes particularly those in enzyme-catalyzed reactions [8]. Hydrogen-bond also plays role in biological systems arising from a stronger directional interaction and to dynamic feature of the proton. It therefore acts as an active site for initiation of chemical reaction.

It is well known that H-bond in crystals of organic compounds often acts as a decision factor governing packing and it helps in the design of novel and interesting crystal structures especially in the area of crystal engineering [9]. Additionally, if one goes by the Pauling [10] definition of hydrogen bonding that: "A hydrogen-bond is an interaction that directs the association of a covalently bound

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hydrogen atom with one or more other atoms, groups of atoms, or molecules into an aggregate structure that is sufficiently stable to make it convenient for the chemist to consider it as an independent chemical species'' thus it nicely corresponds to the patterns which are often observed in crystal structures. The cooperative activity of hydrogen-bond is also involved in the molecules with multiple π-bonds as π-electron delocalization and is related with intermolecular and intramolecular H-bonds [11].

Theoretical studies on several Schiff base derivatives; phenols and their crystal structure have been investigated. The crystal structure of 2-hydroxyacetophenone indicates that the proton is at the hydroxylic oxygen where as in *ortho*-hydroxyacetonapthoimine, proton resides at the nitrogen atom [12]. Additionally, fabrication of artificial receptors through H-bonding, CH-π interaction, π–π stacking along with hydrophobic interaction put their effects as consolidated factors in effective and specific recognition at molecular level [13]. Participation of both H-bonding and π–π stacking interactions has offered rational approach for the understanding of higher order association especially between host and guests [14].

Based on these precedence and also our interest in the design of several conjugated organic compounds for their spectral and bioactivities by complexation especially with Ru^{II}/Ru^{III} ions, we have carried out single-crystal X-ray crystallographic studies of organic compounds like 3-[4-(3-oxo-3-pyridin-2-yl-propenyl)-phenyl]-1-pyridin-2-yl-propenone (L^1), *p*-cholorophenyldiazopentane-2,4-dione (L^2) and *p*-methylphenyldiazopentane-2,4-dione (L^3). The details of the possible structural motifs resulting from their self-association through non-covalent interaction have been reported.

2. Experimental

2.1. Materials and methods

All the chemicals are purchased from Sigma–Aldrich and are used without further purification. Elemental analysis and FAB mass data are recorded on Carbo-Erba elemental analyzer 1108 and JEOL SX-102 mass spectrometer, respectively. IR spectra in the 400–4000 cm^{-1} range were recorded as KBr pellets on a JASCO FT-IR 5300 spectrometer where as ^1H NMR spectra were recorded on a JEOL AL 300 MHz spectrometer using $CDCl_3$ and DMSO-d_6 as solvents and TMS as internal reference.

2.2. Synthesis

L^1 was synthesized using reported procedure [15] by the condensation of 2-acetyl pyridine (0.02 mol, 2.2 ml) with terepthalaldehyde (0.01 mol, 2.2 ml) in alkaline medium. The resulting yellow microcrystalline solid (~80%) melted at 148–150 °C and gave satisfactory elemental analysis and mass ($m/z = 340$). IR (KBr pellet, cm^{-1}) 1670 υ(C═O), 1600 υ(phenyl); ^1H NMR (DMSO-d_6, 300 MHz) δ (ppm) 8.4 (d, 2H; HC═CH), 8.2 (d, 2H; HC═CH), 7.7–8.0 (m, 8H; pyridyl protons), 7.5 (s, 2H; phenyl), 7.4 (s, 2H; phenyl).

L^2 prepared by the general procedure [14] in which *p*-chloroaniline (purchased from Sigma–Aldrich), 0.64 g; were dissolved in 1 N HCl (25 cm^3) at 0–5 °C temperature and cooled aqueous solution (10 cm^3) of $NaNO_2$ (0.40 g) was added drop wise with stirring followed by the addition of pentane-2,4-dione (0.50 g) and sodium acetate (5.0 g) dissolved in water (30 ml). Corresponding mixture was further stirred for 4 h at room temperature (~25 °C). Solids thus obtained were filtered and washed several times with distilled water followed by ethanol and then dried *in vacuo*. The crude product was then re-crystallized in ethanol, which melted at 78–80 °C and showed satisfactory elemental analysis. IR (KBr pellet, cm^{-1}): 1668 υ(C═O), 3070 υ(NH), 1626 υ(C═N); ^1H NMR ($CDCl_3$, 300 MHz) δ (ppm) 14.69 (s, 1H; NH), 7.35 (m, 4H; phenyl), 2.60 (s, 3H; CH_3), 2.48 (s, 3H; CH_3).

L^3 was also prepared using the same procedure as used for L^2, mp. 80–82 °C, and shows satisfactory elemental analysis. IR (KBr pellet, cm^{-1}) 1668 υ(C═O); 1626 υ(C═N); 1587 υ(phenyl); ^1H NMR (300 MHz, $CDCl_3$, ppm) δ 14.8 (s, 1H, —NH); 7.4 (m, 4H, aromatic), 1.8–2.2 (m, 9H, Ar—CH_3 and OC—CH_3); ^{13}C NMR (75 MHz, $CDCl_3$) δ (ppm) 197.4, 197.0 (C═O); 139.2, 135.9, 132.9, 130.2 (phenyl); 116.2 (C═N), 31.6, 26.6 (CO—CH_3); 20.9 (ph—CH_3).

2.3. X-ray crystallography

The X-ray data were collected at 297 K using Cu-Kα radiation ($\lambda = 1.54184$) and graphite monochromator. The structure was solved by direct methods. All non-hydrogen atoms were refined anisotropically by full-matrix least squares. Computations were carried out on a PC using SHELXL-97 [16] program package.

3. Results and discussion

To look into the structural pattern obtained after association of conjugated organic compounds, the chalcone a class of organic compound used as precursor in the synthesis of bioactive flavone derivatives [17] was targeted first in the present study. The compound was crystallized from ethanol:chloroform (1:1) with space group $P2_1/c$. Major crystallographic data are summarized in Table 1. The ORTEP view of the compound is shown in Fig. 1. The bond length between C1—C2 = 1.373(3) Å is quite close to that obtained for aromatic ring system. Other bond length data are in normal length. However, bond between C5—C6 = 1.504(3) Å is slightly longer than the single bond between C6—C7 = 1.474(3), C7—C8 = 1.342(3) Å, is longer than typical double C═C bond (as for example for ethene). Thus these, observations are indicative of partial delocalization in this region of the structure. However,

Table 1
Summary of crystallographic data for L^1, L^2 and L^3

Parameters	L^1	L^2	L^3
Formula	$C_{22}H_{16}N_2O_2$	$C_{11}H_{11}ClN_2O_2$	$C_{12}H_{14}N_2O_2$
M	340.37	238.67	218.27
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/c$	C_2/c	$P2_1/c$
a (Å)	3.8670(2)	18.9160(6)	13.151(3)
b (Å)	12.5840(6)	4.3200(2)	4.7288(10)
c (Å)	17.0560(8)	28.0660(8)	17.895(4)
β (°)	96.356(3)	99.835(2)	103.568(3)
V (Å^3)	824.88(7)	2259.77(14)	1081.8(4)
Z	2	8	2
D_c (mg m^{-3})	1.370	1.403	1.012
Reflns. collected	7127	10,090	6601
Reflns. unique	1519	2108	2633
R_{int}	0.033	0.031	0.046
Index ranges	$-4 \leqslant h \leqslant 4$	$-23 \leqslant h \leqslant 23$	$-17 \leqslant h \leqslant 12$
	$-12 \leqslant k \leqslant 15$	$-5 \leqslant k \leqslant 5$	$-6 \leqslant k \leqslant 6$
	$-21 \leqslant l \leqslant 21$	$-34 \leqslant l \leqslant 34$	$-22 \leqslant l \leqslant 23$
Refinement method	Full-matrix, least squares on F^2		
wR_2	0.1566	0.1603	0.0829
R_1	0.0622	0.0567	0.0520
GoF	1.107	1.071	1.006

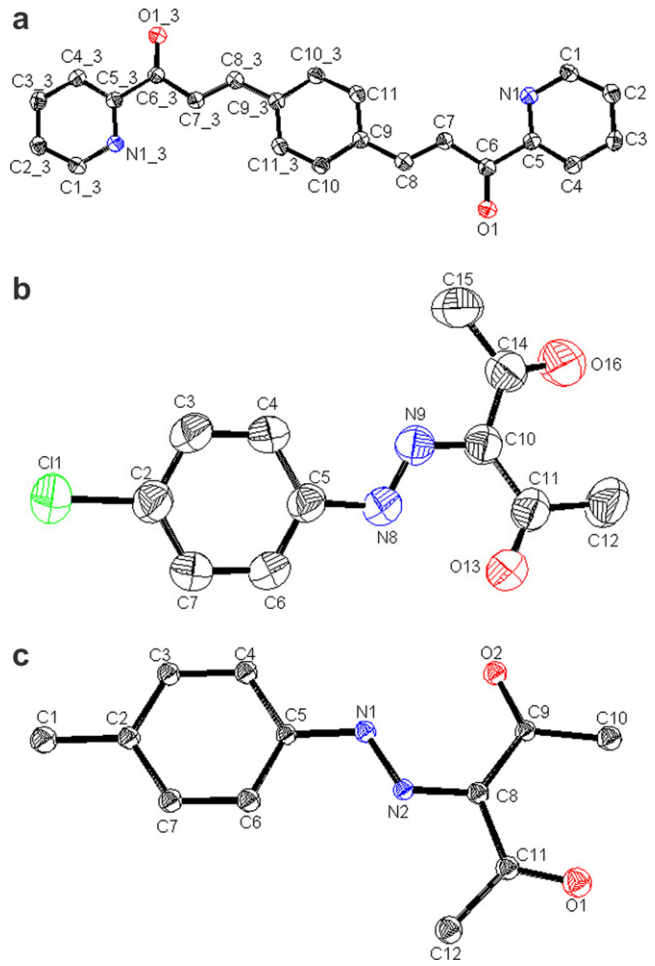

Fig. 1. ORTEP diagram of L^1 (a), L^2 (b) and L^3 (c). Thermal ellipsoids are at 50% probability.

due to highly conjugated nature of the compound, no significant difference in bond radii as compared to the normal one could be observed.

From the X-ray data it is found that the pyridine plane deviates by ±5.0° from the plane of central aromatic ring. The plane of pyridyl ring deviates from the plane containing —C=O group by 0.3°. Thus this structure indicates partial delocalization of electrons in this region in support with the variation of bond lengths between C5—C6, C6—C7 and C7—C8. It can lead to the formation of intermolecular bonds with either complementary charge on the partners or with the complementary functionality present on the partners through non-covalent interactions as reported for several Schiff bases and for benzene-dimer [17]. The intermolecular association between similar molecules via C—H···N (pyridyl) that is CH group lying closer to pyridyl N in L^1 molecule is allowed to interact with *N*-pyridyl of other molecule and CH of other molecule with *N*-pyridyl of earlier molecule results into a single helical motif, shown in Fig. 2. Two equivalent C—H···N hydrogen bonds exist since pyridyl rings are related through inversion centre. According to the Etter's graphs' terminology such a ring may be designated as $R^2_2(6)$ [18].

These $R^2_2(6)$ motifs are observable in crystal structures. The Cambridge Structural Database (CSD) [19] was searched through such patterns existing between pyridyl rings with the reservations that no errors for the crystal structures, not polymeric, no ions and no powder structures exist. Additionally, the conditions for the accuracy of the crystal structures were included, R-factor less than 8% and e.s.d.'s for CC bond lengths less-equal 0.005 Å; X—H bonds were normalized to the mean neutron diffraction corresponding bond lengths according to the rules applied in CSD. Two hundred motifs mentioned above were found, thus centro symmetric dimer with two equivalent C—H···N bonds between pyridyl rings exist sometimes in crystal structures of organic compounds. The histogram of H···N distances within C—H···N hydrogen bonds is presented in Fig. 3. The range of H···N distances amounts to 0.822 Å, the shortest H···N distance is equal to 2.401 Å, the longest one 3.223 Å, mean H···N value – 2.669 Å. For the L^1 structure analyzed here H···N distance is equal to 2.635 Å.

Such associations observed for L^1 are nicely in line with the rules given early on by Etter [18] who stated that:

- all good proton donors and acceptors are used in hydrogen bonding,
- six-membered-ring intramolecular hydrogen bonds form in preference to intermolecular hydrogen bonds,

Fig. 2. Helical assembly (wire form) in L^1 through N(1)···H—C(2) and N(2)···H—C(1) (1 and 2 refer to two molecules).

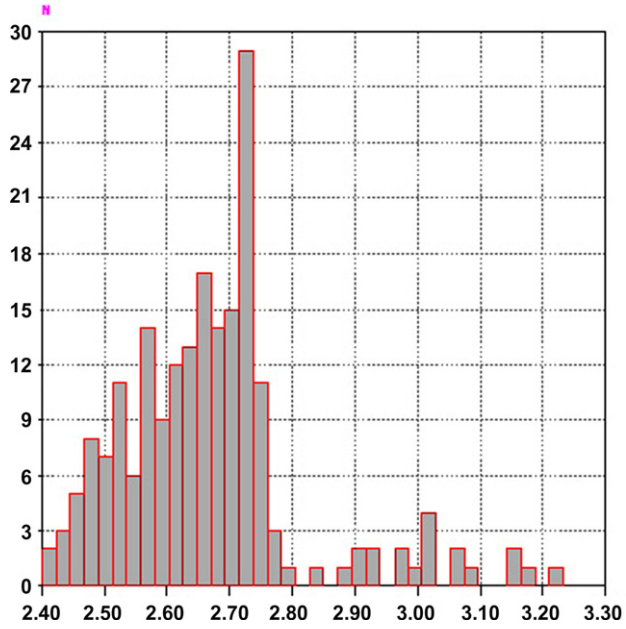

Fig. 3. The histogram of H···N distance (in Å) based on the CSD search and concerning C—H···N bonds in centrosymmetric dimers of pyrroles.

- the best proton donors and acceptors remaining after intramolecular hydrogen-bond formation form intermolecular hydrogen bonds to one another.

Best proton donors and acceptors were defined early on by Donohue [20] and good proton donors are those in carboxylic acids, amides, ureas, anilines, imides and phenols, less acidic protons in acetylenes, aldehydes, activated aromatic and aliphatic compounds while good proton acceptors in acid and amide carbonyl groups, sulfoxides, phosphoryls, nitroxides and amine nitrogens.

One can see that there is the preference in crystal structures to form six-membered rings closed through intramolecular hydrogen bonds. This is nicely shown in the next sections of this study where L^2 and L^3 structures are analyzed.

Etter also stated [18] that if all best proton donors and acceptors are involved in hydrogen bonds, the less acidic and less basic centers are consecutively involved in such interactions. This is visible for the crystal structure of L^1 where there are two typical proton acceptors: oxygen atom of carbonyl group and nitrogen of pyridyl ring. However, there are not any good proton donors. Thus C—H bonds are involved in hydrogen bonds, the motifs containing C—H···N H-bonds were described above and there are also C3—H3···O1 contacts with H···O distance of 2.551 Å which may be hardly classified as hydrogen bonds; for the latter interaction the O-atom of carbonyl group acts as a proton acceptor.

Hence consequently, because of the lack of typical proton donors and only two proton acceptors the other types of interactions are realized for the L^1 crystal structure. When pyridyl CH of one molecule was allowed to interact with π-electron sphere of pyridyl ring of another similar

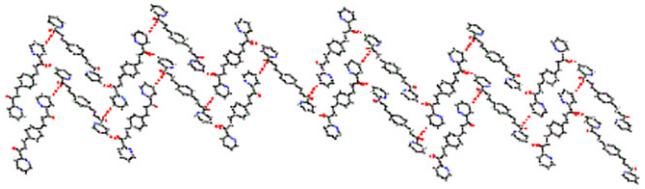

Fig. 4. Double helical structure through C—H···π (2.948 Å).

molecular as depicted in Fig. 4, it results into a double helical structure with r[CH···π] 2.948 Å, providing continuous formation of the cavity.

It is evident that O=C—CH=CH groups do not lie in the same plane as the aromatic rings are hence, show asymmetric distribution of the electron density in this conjugated system. However, as it was indicated earlier, one can observe the π-electron delocalization of C—C bonds (C5—C6, C6—C7 and C7—C8).

The creation of slight positive and negative charges on the whole skeleton of the molecule is evident. This behaviour can provide an opportunity for similar molecules to get associated via complementary charges viz. +ve part of one molecule can be attached to the −ve end of the other molecule or vice versa. Based on this simple electrostatic model concept, when the association of this molecule through C—H···N interactions was examined, it results in a single helical motif. On the other hand C—H interaction with π-electrons (2.948 Å) of aromatic rings results into a double helical motif with the extension of continuous formation of uniform cavity of dimension 20.97 Å in length and 3.37 Å in width. This is more evident in its space-filling model Fig. 5. Additionally, face-to-face association through π–π interaction separated at (3.37 Å) provides nice parallel-unlimited onward arrangements of pyridyl groups.

L^2 crystallizes in monoclinic crystal system with space group C_2/c. The molecular structure and crystallographic numbering scheme are illustrated in Fig. 1. The unit cell contains eight discrete molecules arranged in the form of two helical chains, which extend all along the lattice. Evidently, there are π-stacking among the molecules and there are two different arrangements; two molecules are paired (analogues to parallel displaced case again like benzene dimer) with an average intermolecular distance of 3.57 Å followed by an assembly of three molecules. Phenyl rings of the adjacent molecules from a pair and trio are stacked

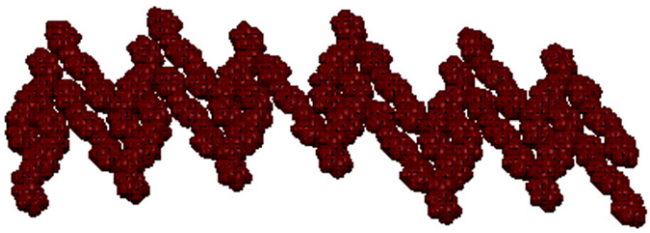

Fig. 5. Space-filled model of double helical structure in L^1 showing formation of continuous cavity.

perpendicularly (the closest C—C intermolecular distance is ~3.484 Å) with their tails in opposite directions.

Each molecule has an almost planar structure as can be expected in any such conjugated system. However, the sp^3 hybridized N8 atom prevails perfect planarity. The inter planar angle between the planes constituted by the phenyl ring and the quasi N_2C_2OH ring ($HN_8N_9C_{10}C_{11}O_{13}$) is 3.7°. Interestingly, both the oxygen atoms are in keto form. There is a double bond between C10 and N9 (bond length is 1.315(3) Å). N9—N8 and N8—C5 are thus single bonds and the conjugation breaks at this point. There is an intramolecular hydrogen bond (N8—H···O13) with N8—O13 of 2.585 Å, which is shorter than the sum of their covalent radii (2.9 Å).

L^3 crystallizes in monoclinic crystal system with space group $P2_1/c$. The molecular structure and crystallographic numbering scheme are illustrated in ORTEP. The unit cell contains two discrete molecules. There is substantial arrangement of molecules through CH···π interaction in one-dimensional forming uniform cavity with length of 7.632 and 3.908 Å width. The distances between phenyl ring and terminal methyl hydrogen lie between 2.780 and 2.866 Å.

Additionally, there are intermolecular as well as intramolecular N···H interactions. Intramolecular N···H (methyl) distances amount to 2.739 Å whereas intermolecular distances are equal H(phenyl)···N 2.460 Å, H(methyl)···N 2.579 Å, H(methyl)···N 2.638 Å.

One can see that for both L^2 and L^3 crystal structures, the six-membered rings closed by N—H···O intramolecular hydrogen bonds are created (Fig. 6). According to the Etter graphs' nomenclature they may be designated as S(6). As it was mentioned earlier there is the preference of the formation of such motifs in crystal structures. Numerous studies were performed on S(6) connections where homonuclear O—H···O hydrogen bonds exist but also N—H···O, N—H···N and O—H···N interactions of S(6)'s type were analyzed. They are usually attributed to so-called resonance assisted hydrogen bonds (RAHBs) [21]. Briefly speaking the concept of RAHBs' systems is based on the statement that owing the existence of conjugated system of single and double bonds the π-electron delocalization exists which causes the enhancement of the strength of hydrogen bonds. Such concept is criticized recently [22] but there is no doubt

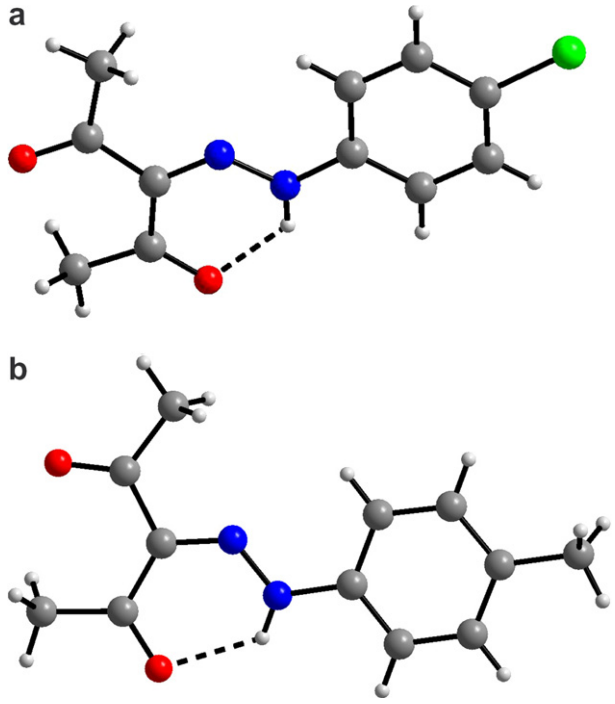

Fig. 6. Creation of six-membered ketohydrazone pseudo-ring in L^2 (a) and L^3 (b).

Table 2
The geometrical parameters (in Å) of six-membered ketohydrazone rings taken from Cambridge Structural Database, mean, minimum and maximum values are also included

Refcode	N=N	C=C	C—C	C=O	H···N	N···O
ADEGAS	1.292	1.319	1.464	1.225	1.754	2.575
BARPAM	1.314	1.309	1.492	1.233	1.789	2.570
CIVYIP01	1.309	1.320	1.493	1.236	1.709	2.538
CIVZEM10	1.315	1.329	1.487	1.241	1.784	2.565
CIVZEM10	1.315	1.315	1.501	1.241	1.769	2.601
DIRQEA	1.300	1.330	1.453	1.229	1.747	2.596
DIRQIE	1.299	1.319	1.466	1.224	1.772	2.560
DIRQUQ	1.309	1.299	1.474	1.211	1.795	2.615
HENJOZ	1.301	1.327	1.470	1.234	1.795	2.559
HENJUF	1.304	1.327	1.472	1.232	1.790	2.561
JARPEX	1.307	1.348	1.454	1.284	1.691	2.556
JARPEX01	1.309	1.339	1.455	1.262	1.746	2.553
KAQSAW	1.305	1.333	1.458	1.257	1.655	2.502
LALQIZ	1.311	1.317	1.478	1.226	1.755	2.542
LEZHON	1.295	1.341	1.456	1.273	1.627	2.489
LIKFUG	1.341	1.329	1.497	1.259	1.786	2.569
NAMMEU	1.308	1.337	1.457	1.262	1.705	2.548
NAMMEU01	1.307	1.336	1.459	1.265	1.707	2.548
NAMMEU02	1.305	1.340	1.454	1.268	1.679	2.545
NAMMEU03	1.303	1.344	1.447	1.276	1.669	2.541
OLOCAT	1.301	1.346	1.458	1.268	1.595	2.485
PAMBOO01	1.306	1.342	1.437	1.276	1.721	2.539
PAMBOO01	1.305	1.346	1.440	1.276	1.749	2.548
SANZEM	1.304	1.316	1.501	1.231	1.781	2.579
SCNPHO	1.339	1.311	1.475	1.239	1.762	2.517
TAPFEV	1.306	1.330	1.464	1.235	1.768	2.586
TEJJUO	1.298	1.309	1.490	1.205	1.793	2.599
UNEWUF	1.313	1.310	1.500	1.236	1.733	2.562
UNEWUF01	1.313	1.308	1.515	1.233	1.754	2.560
UNEWUF01	1.311	1.306	1.500	1.237	1.717	2.571
VEHXOV	1.300	1.329	1.466	1.260	1.767	2.546
WIKWOC	1.301	1.341	1.458	1.265	1.702	2.519
WIKWUI	1.290	1.340	1.461	1.270	1.649	2.500
WIKXAP	1.303	1.338	1.460	1.270	1.629	2.530
YAPDID	1.313	1.332	1.474	1.282	1.688	2.542
YAWVUN	1.311	1.324	1.475	1.232	1.744	2.575
YAWVUN01	1.311	1.325	1.477	1.239	1.790	2.579
YAWWIC	1.293	1.336	1.464	1.234	1.771	2.565
ZUCQOD	1.298	1.338	1.442	1.264	1.662	2.514
ZUCRAQ	1.299	1.342	1.433	1.281	1.705	2.540
Mean value	1.307	1.328	1.469	1.249	1.730	2.552
Minimum	1.290	1.299	1.433	1.205	1.595	2.485
Maximum	1.341	1.348	1.515	1.284	1.795	2.615

that the π-electron delocalization is observed for RAHBs and that in extreme cases one can observe the equalization of bonds within the pseudo-ring with the nearly centre position of hydrogen atom. The crystal structure of benzylacetone where S(6) motif with intramolecular O—H···O hydrogen bond is an example of the latter situation [23].

For L^2 and L^3 structures analyzed here the ketohydrazone pseudo-ring with N—H···O bond is observed. Such ketohydrazone species were analyzed in detail previously experimentally since numerous crystal structures were reported and also theoretically since DFT calculations on the model species were carried out [24]. We have performed the search through Cambridge Structural Database to find ketohydrazone rings with intramolecular N—H···O hydrogen bonds. The same reservations and conditions concerning the accuracy of the determination of crystal structures' measurements were applied here as for the C—H···N bonds between the pyridyl rings analyzed in the previous section.

Table 2 presents the geometrical parameters of six-membered ketohydrazone pseudo rings closed through intramolecular N—H···O hydrogen bonds. Forty such rings were searched in CSD which fulfill the accuracy and the other criteria mentioned above. The shortest N···O distance given in Table 2 is equal to 2.485 Å and the mean value amounts to 2.552 Å, this indicates that intramolecular N—H···O hydrogen bonds of ketohydrazones belong to strong ones. One can see that the geometrical parameters of L^2 and L^3 investigated here are well fixed into the ranges presented in Table 2.

4. Conclusion

Two classes of simple organic compounds bearing functional groups on the conjugated structural frame are prepared. These structures have been exploited for the generation of novel single and double helical structural motifs upon the self-association through non-covalent interactions. For all crystal structures analyzed herewith, the typical intra and intermolecular motifs were found, these are for example $R^2_2(8)$ between pyridyl rings and S(6) of intramolecular hydrogen bond.

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Appendix A. Supplementary data

CCDC Reference Nos. 297270, 244360 and 619000 contains the supplementary crystallographic data for L^1, L^2 and L^3, respectively. These data can be obtained free of charge via http://www.ccdc.cam.ac.uk/conts/retrieving.html, or from the Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK, fax: +44 1223 336 033, or e-mail: deposit@ccdc.cam.ac.uk. Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.molstruc.2007.08.011.

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