Crystal Structure of 3,4,5,6-Tetrahydrophthalic Anhydride at 150 K

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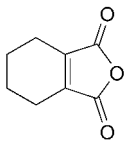
The crystal structure of 3,4,5,6-tetrahydrophthalic anhydride, (=4,5,6,7-tetrahydroisobenzofuran-1,3-dione; **1**; $C_8H_8O_3$) was determined and refined by an analysis of three-dimensional X-ray-diffraction data at 150 K. This bicyclic compound crystallizes in space group *Pbca* with two symmetry-independent molecules **I** and **II** per asymmetric unit. The cyclohexene ring in both molecules adopts a half-chair conformation. The obtained conformational descriptions of the six-membered rings in the crystal phase are consistent with conformational data derived from molecular-orbital calculations. The structure analysis evidences considerable distorsion of the partially hydrogenated six-membered ring; the furan ring is flattened in molecule **I** and slightly deviated from planarity in molecule **II**. The short intermolecular distances found for C=O ··· C=O are interpreted as evidence for nonbonded interactions of the dipole – dipole type. The rather long O ··· H distances indicate that the C(sp^3)-H ··· O interactions are weak.

Introduction. – Within the context of studies on the conformations of bicyclic anhydrides, the structure of *cis*-1,2,3,6-tetrahydrophthalic anhydride $C_8H_8O_3$ has been determined by X-ray diffraction [1]; its crystal structure consists of two symmetry-independent molecules per asymmetric unit. Their conformations are folded, both possessing a boat form for the cyclohexene ring, in agreement with experimental NMR solution data [2][3]. Crystal packing of the two independent molecules in the unit cell has also been investigated [4], establishing non-bonded interactions of the dipole–dipole type between local electrical dipoles of the carbonyl groups C=O ··· C=O. Concomitantly, several techniques of semi-empirical and *ab initio* molecular-orbital calculations have been applied for the determination of the molecular geometry of this compound and its isomers, 3,4,5,6-, 1,2,5,7-, and 1,4,5,6-tetrahydrophthalic anhydrides [5]. These calculations confirmed the preference of the folded conformation for *cis*-1,2,3,6-tetrahydrophthalic anhydride and revealed large differences in the stereochemical behavior of these isomeric tetrahydrophthalic anhydrides. In view of the considerable interest in the structural properties of this series of anhydride isomers, it appeared desirable to have precise structural data for a selected compound to provide a check of the theory and possibly to aid in formulating refinements in the calculations. Experimental structures of the gas-phase molecules have not yet been reported, and, besides two crystallographic studies on derivatives of 3,4,5,6-tetrahydrophthalic anhydride [6][7], no experimental data on the parent compound are available. Thus, in the present work, an investigation of the crystal structure of 3,4,5,6-tetrahydrophthalic anhydride (=4,5,6,7-tetrahydroisobenzofuran-1,3-dione; **1**) was undertaken to reveal the most stable conformation and to elucidate the structural differences between

the 3,4,5,6-tetrahydrophthalic and *cis*-1,2,3,6-tetrahydrophthalic isomers. Because the tetrahedral C-atoms exhibit unusually large anisotropic displacement parameters without apparent statistical disorder and interatomic distances shorter than the normal C–C bonds at room temperature [8], a low-temperature structure determination was performed. The results of such an analysis for a single crystal of **1** at 150 K led to an improved agreement of the crystal-structure model and the experimental data of the related compounds.



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Results and Discussion. – The asymmetric unit of the single crystal of **1** contains two symmetry-independent molecules **I** and **II** as has been shown for the crystal of the *cis*-1,2,3,6-tetrahydrophthalic isomer [1]. The structures of the two independent molecules are very similar (*Fig. 1*). As implied by the semiquantitative rules of *Bucourt* for conformational analysis [9][10], the cyclohexene ring is assumed to exist in a distorted half-chair form in the two independent molecules. This typical conformation of the six-membered ring observed for **1** has also been established to be the preferred one by geometry-optimized AM1, PM3, and MNDO semi-empirical methods and *ab initio* calculations at both the RHF/PS-31G* and MP2/PS-31G* levels by using the gradient method [5]. The present results obtained for the crystal phase of **1** add additional support to the conformational data derived from IRC calculations by using the PM3 semi-empirical method, which indicate the presence of two conformational minima for **1** (*Fig. 2,a*), both possessing a half-chair conformation for the cyclohexene ring (*Fig. 2,b*). This significant difference from the conformational preference of the cyclohexene ring in the crystal of the *cis*-1,2,3,6-tetrahydrophthalic isomer was expected. The dissimilarities may be attributed mainly to the difference in the position of the C=C bond in the bicyclic system. The degree of freedom between the bonds in the cyclohexene ring is more restricted in *cis*-1,2,3,6-tetrahydrophthalic anhydride owing to the *cis* orientation of the furan ring with respect to the cyclohexene ring [11].

A comparison of structural parameters between the two independent molecules **I** and **II** shows internal consistency. The most-prominent discrepancies are displayed among the parameters determining the geometry of the cyclohexene ring, which shows significant deviations from a C_2 symmetric internal top. The ring dihedral angles at the bonds C(7)–C(6), C(6)–C(5), and C(4)–C(5) in molecule **I** (absolute values 16.8(3), 46.2(2), and 61.5(2)°, resp.) are larger than the corresponding angles in molecule **II** (absolute values 13.1(3), 42.9(2), and 61.3(2)°, resp.) (*Table 1*)[1]). For the C(2)–C(3) and C(3)–C(4) bonds, the absolute values of the dihedral angles of **I** (11.6(3) and 41.1(2)°, resp.) are both lower than those observed in **II** (14.5(3) and 44.1(2)°, resp.). All the dihedrals indicated above deviate significantly from the values of unfused cyclohexene rings [12] (absolute values 15.2, 44.9, and 60.2° at the bonds $C(sp^2)-C(sp^3)$

[1]) Arbitrary numbering, according to *Fig. 1*.

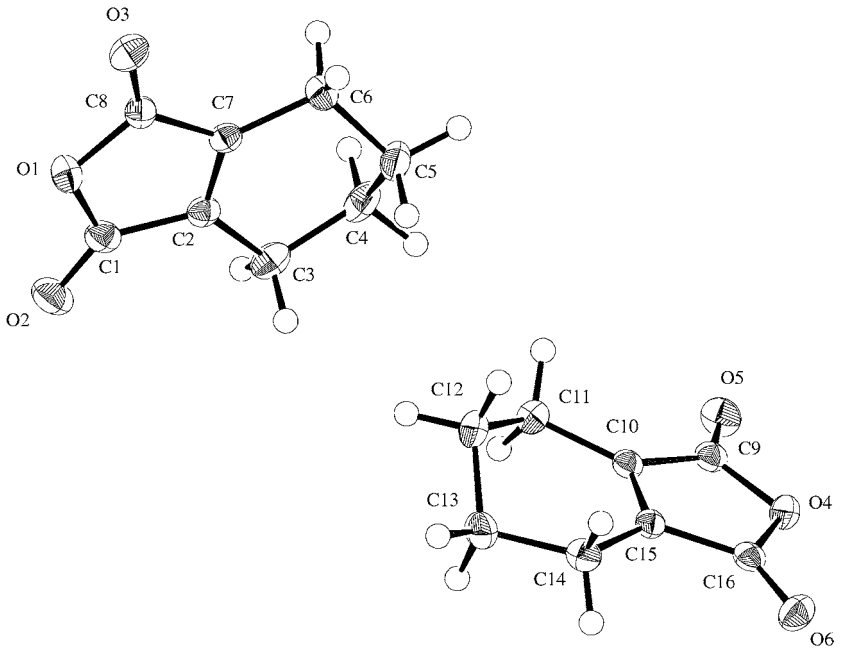

Fig. 1. *ORTEP View of the molecules* **I** *and* **II** *in the asymmetric unit, showing the labelling of the non-H-atoms*[1]). Ellipsoids are drawn at the 50% probability level.

(C=C), $C(sp^3)-C(sp^3)$ (C=C), and $C(sp^3)-C(sp^3)$, resp.), which adopt an ideal half-chair conformation. We may assume that deviations from these values are an index of strain with respect to the undistorted cyclohexene derivatives, and that the differences represent the angular deformations from the ideal half-chair conformations. Also, in both independent molecules **I** and **II**, the torsional angles deviate significantly from the corresponding values calculated for the two conformers of **1** at the MP2/PS-31G* level [5] (absolute values 14.6, 44.3, and 63.0° at the bonds $C(sp^2)-C(sp^3)$ (C=C), $C(sp^3)-C(sp^3)$ (C=C), and $C(sp^3)-C(sp^3)$, resp.). The distortion of the cyclohexene ring in **1**, owing to the fusion with the furan ring, could not be detected theoretically at the *ab initio* MP2 level as well as at different levels of semiempirical molecular-orbital calculations [5]. This indicates that the distortion of the cyclohexene rings detected in the crystal phase is undoubtly due to crystal-packing forces and, as will be discussed later, the differences in the changes in the chemically equivalent parts of the cyclohexene rings are nevertheless consistent with packing effects. As far as the valence angles are concerned, very slight differences are found in some intra-cyclohexene ring angles such as C(3)–C(2)–C(7) and its chemical equivalent C(2)–C(7)–C(6), which deviate by 0.6(2) and 0.5(2)°, respectively, in molecule **II** compared to molecule **I**. These differences are obviously due to packing effects since there is no chemical difference between the two molecules. The angles within the furan ring in both molecules are normal compared to those observed in maleic anhydride [13] and some of its derivatives [6][7], which exhibit similar deviations from the data of succinic

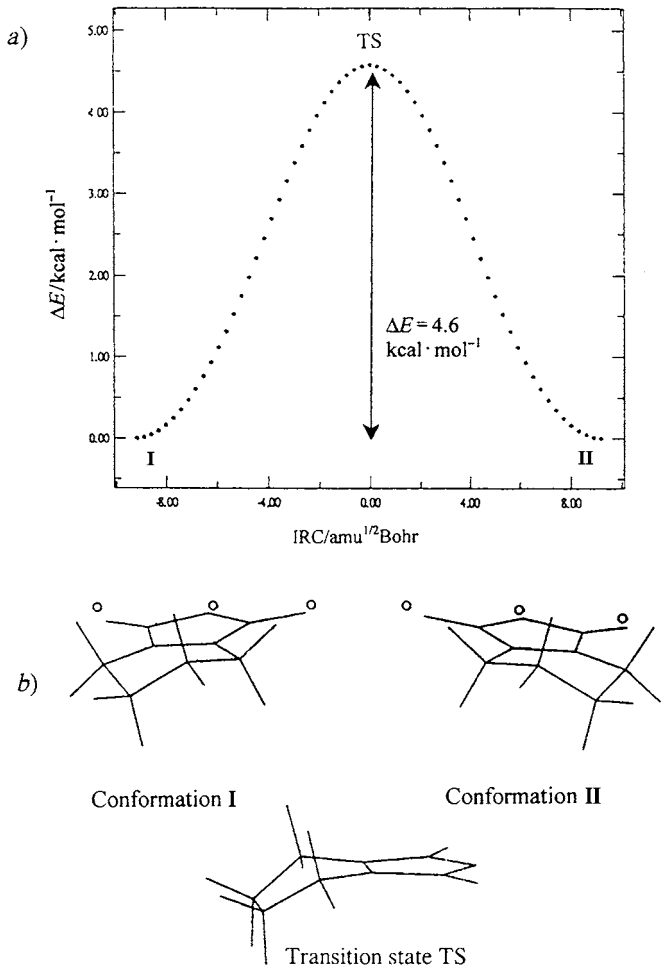

Fig. 2. a) *Energy profile of 3,4,5,6-tetrahydrophthalic anhydride* (**1**) *obtained from IRC by the PM3 semiempirical method.* b) *Calculated ground-state conformations* **I** *and* **II** *of* **1** *and the structure of the transition state* (TS).

anhydride [14] and *cis*-1,2,3,6-tetrahydrophthalic anhydride [1]. The geometry of the furan ring in molecule **I** is planar or nearly so: all of the ring dihedral angles are less than or equal to 0.2(2)° in absolute values. In molecule **II**, however, the ring dihedrals are in the range 0.9(2)–2.9(2)° with absolute values of 2.3(2), 2.9(2), and 2.4(3)° at the bonds C(10)–C(9), O(4)–C(9), and C(16)–O(4), respectively. Accordingly it seems appropriate to describe the geometry of molecule **II** as slightly nonplanar, being puckered at the O(4) atom. The C–O and C=O bond lengths as well as the C–C bond length in the furan ring of the two independent molecules agree well with those found in maleic anhydride [13] and its derivatives [6][7]. The C=C bond distance (1.331(2) Å in **I** and 1.332(2) Å in **II**) is in good agreement with those observed in 3,4,5,6-

Table 1. *Selected Geometric Parameters for 3,4,5,6-Tetrahydrophthalic Anhydride* (**1**)[1]). Esd. are given in parentheses.

Molecule **I**		Molecule **II**	
Bond lengths [Å]: O(1)–C(8)	1.396(2)	Bond lengths [Å]: O(4)–C(9)	1.399(2)
O(1)–C(1)	1.401(2)	O(4)–C(16)	1.396(2)
O(2)–C(1)	1.193(2)	O(5)–C(9)	1.193(2)
O(3)–C(8)	1.192(2)	O(6)–C(16)	1.193(2)
C(1)–C(2)	1.472(2)	C(16)–C(15)	1.474(3)
C(2)–C(7)	1.331(3)	C(15)–C(10)	1.332(3)
C(7)–C(8)	1.474(3)	C(9)–C(10)	1.480(3)
C(2)–C(3)	1.487(3)	C(14)–C(15)	1.487(3)
C(3)–C(4)	1.526(3)	C(13)–C(14)	1.528(3)
C(4)–C(5)	1.527(3)	C(12)–C(13)	1.531(3)
C(5)–C(6)	1.528(3)	C(11)–C(12)	1.530(3)
C(6)–C(7)	1.487(3)	C(10)–C(11)	1.490(3)
Valence angles [°]:		Valence angles [°]:	
C(1)–O(1)–C(8)	107.5(1)	C(9)–O(4)–C(16)	107.6(1)
C(8)–C(7)–C(2)	108.5(2)	C(9)–C(10)–C(15)	108.0(2)
O(2)–C(1)–O(1)	120.3(2)	O(6)–C(16)–O(4)	121.0(2)
C(7)–C(6)–C(5)	108.8(2)	C(10)–C(11)–C(12)	109.3(2)
C(6)–C(5)–C(4)	112.1(2)	C(11)–C(12)–C(13)	112.3(2)
C(4)–C(3)–C(2)	109.8(2)	C(13)–C(14)–C(15)	109.6(2)
C(3)–C(2)–C(7)	125.4(2)	C(14)–C(15)–C(10)	124.8(2)
C(2)–C(7)–C(6)	124.9(2)	C(11)–C(10)–C(15)	125.4(2)
Dihedral angles [°]:		Dihedral angles [°]:	
O(1)–C(8)–C(7)–C(2)	– 0.2(2)	O(4)–C(9)–C(10)–C(15)	– 2.3(2)
C(8)–C(7)–C(2)–C(1)	0.2(2)	C(9)–C(10)–C(15)–C(16)	0.9(2)
O(1)–C(1)–C(2)–C(7)	– 0.1(2)	O(4)–C(16)–C(15)–C(10)	0.9(2)
C(2)–C(1)–O(1)–C(8)	– 0.1(2)	C(9)–O(4)–C(16)–C(15)	– 2.4(2)
C(3)–C(2)–C(7)–C(6)	– 0.6(3)	C(11)–C(10)–C(15)–C(14)	1.2(3)
C(2)–C(7)–C(6)–C(5)	– 16.8(3)	C(12)–C(11)–C(10)–C(15)	13.1(3)
C(3)–C(4)–C(5)–C(6)	– 61.5(2)	C(11)–C(12)–C(13)–C(14)	61.3(2)
C(2)–C(3)–C(4)–C(5)	41.1(2)	C(12)–C(13)–C(14)–C(15)	– 44.1(2)
C(4)–C(3)–C(2)–C(7)	– 11.6(3)	C(10)–C(15)–C(14)–C(13)	14.5(3)
C(4)–C(5)–C(6)–C(7)	46.2(2)	C(10)–C(11)–C(12)–C(13)	– 42.9(2)
C(1)–O(1)–C(8)–C(7)	0.2(2)	C(10)–C(9)–O(4)–C(16)	2.9(2)

tetrahydrophthalimide [15] and 8,9,10-trinorborn-2-ene-2,3-dicarboxylic anhydride-$C_9H_8O_3$ [6] (1.324(5) and 1.332(4) Å, resp.), which possess a similar furan-ring moiety.

One of the most interesting features is the crystal packing of **1** (*Fig. 3*). The packing pattern reveals a large number of close intermolecular contacts among the electrophilic C-atoms (El) and their concomitant nucleophilic O-atoms (Nu) of symmetry-related molecules **I** and **II** as well as between the H- and the O-atoms of symmetry-independent molecules **I** and **II** that are within the range of the *Van der Waals* interactions. The significant intermolecular Nu–El C=O···C=O bonds that are within the presently acceptable limit ($d(O\cdots C) \leq 3.4$ Å) [16], the noncovalent angles θ (O···CO), and the dihedral angles φ (O···COC) (CO being a carbonyl group) defining the Nu–El O···C atom abstraction distance are reported in *Table 2*. The minimum intermolecular C=O···C=O distance is 2.957(2) Å between molecules **I** and 2.926(2) Å between molecules **II** (*Fig. 4*). The latter short contacts are significantly smaller than the sum of the *Van der Waals* radii, which is assumed to be 3.2 Å [17]. All

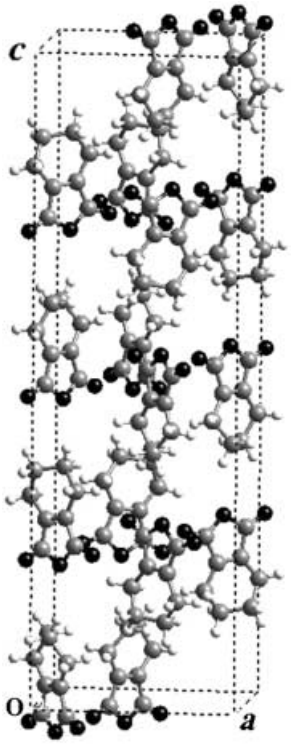

Fig. 3. *View of the structural arrangement of* **1** *down [0 1 0]*. O-Atoms black, C-atoms grey, and H-atoms white.

the distances indicated above and the other ones reported in *Table 2* are within the range of values observed for several contacts of the same type in the crystal structure of *cis*-1,2,3,6-tetrahydrophthalic anhydride [1][4]. They are also in good agreement with the corresponding values found by *Cossu et al.* [16] for a series of mono- and polycyclic anhydride compounds that also exhibit solid-state intermolecular Nu–El C=O···C=O interactions. The values of the noncovalent angles θ (O···CO) are in the range 83.1(1) – 89.5(1)° with only two θ values out of the bisector criteria ($\varphi \approx 90°$) [16]. If we exclude these uncertain interactions, we can state that the carbonyl groups are involved in significant C=O···C=O dipole–dipole interactions. In the two other cases ($\theta = 83.4(1)$ and 83.1(1)°), the nucleophilic O-atom is in close contact with the electrophilic carbonyl group (O(5)···C(16) distance of 3.195(2) Å and O(3)···C(1) distance of 3.191(2) Å; see *Table 2*), and both correspond to carbonyl groups forming dimers held together by somewhat weaker Nu–El dipole–dipole C=O···C=O interactions.

In addition to the Nu – El C=O···C=O interactions, the crystal packing of 3,4,5,6-tetrahydrophthalic anhydride (**1**) is stabilized by weak C(sp^3)–H···O intermolecular interactions [18]. Geometric details of the H-bonds that are within the accepted range (d(O···H) ≤ 2.8 Å and θ (C–H···O) > 120°) are given in *Table 3*. The most-significant intermolecular H-bonds observed in this molecular packing are C(14)–H(15)···O(5) (2.59(2) Å, 130(2)°) and C(11)–H(9)···O(5) (2.66(2) Å, 161(2)°) between H-atoms of

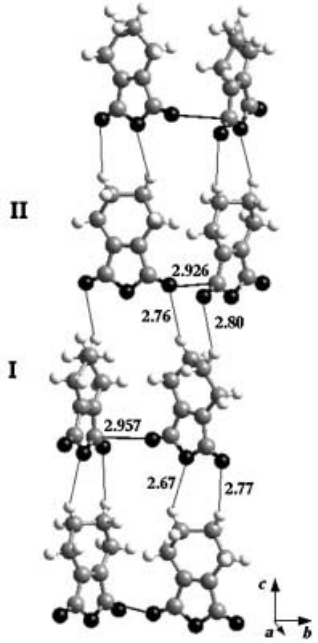

Fig. 4. *Details of some interactions between molecules of 3,4,5,6-tetrahydrophthalic anhydride* (**1**). The figure evidences the shortest contacts C=O···C=O between molecules of the same type **I** or **II** and the H-bonds interconnecting the molecules **I** and **II**.

Table 2. *Geometric Parameters* (d, θ and φ) *of the Intermolecular C=O···C=O Bonds*

C=O···C=O	Type	d(O···C) [Å]	θ (O···CO)[°]	φ (O···COC)[°]
C(1)=O(2)···C(8)=O(3)	**I**–**I**	2.957(2)	89.0(1)	91.8
C(1)=O(2)···C(8)=O(3)	**I**–**I**	3.109(2)	87.2(1)	107.2
C(8)=O(3)···C(1)=O(2)	**I**–**I**	3.148(2)	88.8(1)	71.0
C(8)=O(3)···C(1)=O(2)	**I**–**I**	3.191(2)	83.4(1)	66.9
C(16)=O(6)···C(9)=O(5)	**II**–**II**	2.926(2)	89.5(1)	100.2
C(9)=O(5)···C(16)=O(6)	**II**–**II**	3.195(2)	83.1(1)	115.1

the six-membered ring and the carbonyl O-atom of molecules **II**. The shortest intermolecular C(sp^3)–H···O H-bonds observed between molecules **I** are C(3)–H(1)···O(3) (2.71(2) Å, 129(2)°) and C(6)–H(7)···O(1) (2.79(2) Å, 155(2)°) involving both the O(3) and O(1) atoms of the anhydride moiety. Further analysis shows that symmetry-independent molecules **I** and **II** are arranged into infinite chains along the *c*-axis through four weak H-bonds: C(13)–H(13)···O(1) (2.67(2) Å, 141(2)°), C(12)–H(12)···O(3) (2.77(2) Å, 153(2)°), C(4)–H(3)···O(6) (2.76(2) Å, 157(2)°), and C(5)–H(6)···O(5) (2.80(2) Å, 132(2)°) (*Fig. 4)*. These chains are connected in the lateral directions by more-significant interactions between symmetry-dependent molecules (type **I**–**I** or **II**–**II**), as specified in *Table 2* for the C=O···C=O interactions and in *Table 3* for the C–H···O H-bonds.

Clearly, the strong acceptor ability of the carbonyl O-atoms dominates the packing of 3,4,5,6-tetrahydrophthalic anhydride (**1**) and its isomer *cis*-1,2,3,6-tetrahydrophthalic anhydride [1][4]. Both structures contain C=O···C=O and C–H···O

Table 3. *Geometric Parameters* (d and θ) *of the Intermolecular C(sp³)–H···O Hydrogen Bonds*

C–H···O	Molecules	d(H···O) [Å]	θ(C–H···O)[°]
C(3)–H(1)···O(3)	**I**–**I**	2.71(2)	129(2)
C(4)–H(3)···O(6)	**I**–**II**	2.76(2)	157(2)
C(5)–H(6)···O(5)	**I**–**II**	2.80(2)	132(2)
C(6)–H(7)···O(1)	**I**–**I**	2.79(2)	155(2)
C(11)–H(9)···O(5)	**II**–**II**	2.66(2)	161(2)
C(11)–H(10)···O(6)	**II**–**II**	2.76(2)	127(2)
C(12)–H(12)···O(3)	**II**–**I**	2.77(2)	153(2)
C(13)–H(13)···O(1)	**II**–**I**	2.67(2)	141(2)
C(14)–H(15)···O(5)	**II**–**II**	2.59(2)	130(2)

Table 4. *Crystal Data, Experimental Conditions, and Details of Structure Refinement*

Empirical formula	$C_8H_8O_3$
Formula weight [g·mol^{-1}]	152.15
Temperature [K]	150
Crystal system	Orthorhombic
Space group	*Pbca*
Unit-cell dimensions:	
a [Å]	9.7936(2)
b [Å]	9.0338(1)
c [Å]	31.7069(6)
$\alpha=\beta=\gamma$ [°]	90
V [Å^3]	2805.22(7)
Z	16
Calc. density D_x [Mg·m^{-3}]	1.441
Crystal size [mm]	0.68 × 0.34 × 0.31
Radiation, wavelength [Å]	AgK_α, 0.5608
Absorption coefficient μ [mm^{-1}]	0.068
Measurement method	φ and ω scans
Bragg-angle limits [°]	3.0–20.0
Limiting indices	−11→11, −10→10, −38→29
Measured reflections	13803
Independent reflections	2535
Observed reflections	1882 with $I>2\sigma(I)$
Number of variables	264
Final agreement factors	$R=0.038$, $wR=0.044$
Goodness-of-fit on F	$S=1.44$
Largest peak and hole in the last difference-*Fourier* synthesis [e Å^{-3}]	0.24, −0.23

bonds, together with interactions involving the carbonyl O-atom and either C(sp^3)–H or C(sp^2)–H (in *cis*-1,2,3,6-tetrahydrophthalic anhydride). As pointed out in our structure analysis of *cis*-1,2,3,6-tetrahydrophthalic anhydride [1][4] and its derivatives [16], an obvious relationship between the postulated typical intermolecular interactions and the packing modes of **1** can be predicted. The study of the efficiency of these intermolecular interactions in inducing geometrical deformation of the structure of **1** is undertaken and the results obtained will be reported in an independent work.

Experimental Part

Synthesis. The 3,4,5,6-tetrahydrophthalic anhydride (**1**) was prepared by complete isomerization of a pure sample of *cis*-1,2,3,6-tetrahydrophthalic anhydride (m.p. 376 K) at 473 K, with P_2O_5 as acid catalyst according to *Bailey* and *Amstutz* [19]. Subsequent purification by recrystallization from anh. cyclohexane yielded pure **1**. M.p. 357 K, ^{1}H-NMR ($CDCl_3$): 1.5 (*m*, 2 CH_2), 2.5 (*m*, 2 CH_2). ^{13}C-NMR ($CDCl_3$): – 56.41 (*s*, 2 CH_2); – 56.27 (*s*, 2 CH_2); 68.12 (*s*, C=C); 87.86 (*s*, 2 C=O).

Colorless crystals were obtained by slow evaporation of a dilute soln. of pure samples of **1** in anh. cyclohexane at r.t. in a dry-box.

X-Ray Structure Determination. As the crystals were hygroscopic, the single crystal used was mounted in a thin-walled glass capillary of the *Lindemann* type during data collection. The intensity data were collected at 150 K on a *Nonius* four-circle diffractometer equipped with a *CCD* bidimensional detector and an *Oxford Cryostream* crystal-cooling system. Crystal data, details of data collection, and structure refinement are summarized in *Table 4*. Intensities were corrected for *Lorentz* and polarization effects. No absorption corrections were applied. The structure was solved by direct methods with the SIR92 program [20] and refined by the full-matrix least-squares method, based on *F*, by using the teXsan software [21]. The refinement was performed with anisotropic temperature factors for all non-H-atoms. H-Atoms were refined isotropically. A correction for secondary extinction [22] was applied with a value of the extinction parameter equal to $1.0(2) \cdot 10^{-6}$. The pictures of the crystal structure are shown in *Figs. 1*, *3*, and *4*. The ORTEPII program [23] was used to produce the asymmetric unit of the structure (*Figs. 1*), while the Cerius2 software [24] was applied to represent the molecular arrangement (*Figs. 3* and *4*). Crystallographic data (excluding structure factors) have been deposited with the *Cambridge Crystallographic Data Centre*, as deposition No. CCDC 223922, and can be obtained *via* http://www.ccdc.cam.ac.uk/conts/retrieving.html (or from the CCDC, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44 1223 336033; e-mail: deposit@ccdc.cam.ac.uk).

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