Structure of Mellitic Trianhydride

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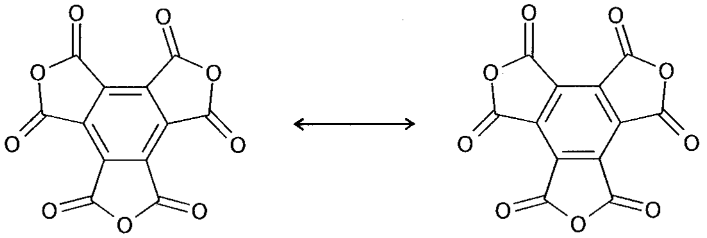
Mellitic trianhydride (MTA; *benzene-1,2:3,4:5,6-hexacarboxylic trianhydride*), a powerful π-electron acceptor, crystallizes in the cubic space group *Pa*3 with four molecules in the unit cell and is orientationally twofold disordered. The molecules are packed exclusively *via* edge-to-face contacts also seen in the isomorphous crystal structures of benzene hexacarbonitrile, benzene hexamine, and all-*trans*-hexachlorocyclohexane. The orthorhombic crystal structure of benzene is also related to these cubic cases, albeit in distorted fashion. The MTA molecules are non-planar in the crystal, taking the shape of a shallow propeller with approximate D_3 symmetry. The non-planarity is ascribed to nucleophilic-electrophilic intermolecular interactions between the five-membered-ring O-atoms and the carbonyl C-atoms. Due to the disorder, the two non-equivalent inner C,C bond lengths of the benzene ring of MTA cannot be resolved. The MTA molecules in the polar crystals of the 1:1 molecular charge-transfer complexes with triphenylene and 9,10-dimethylanthracene are ordered and practically planar, and, within experimental error, show equal inner C,C lengths, despite severe adjacent bond-angle distortions. Accordingly, the structure of MTA provides no evidence in support of a so-called '*Mills-Nixon* effect'.

Introduction. – Mellitic trianhydride (MTA, *benzene-1,2:3,4:5,6-hexacarboxylic trianhydride; Scheme 1*), formally an oxide of carbon ($C_{12}O_9$) [1], is a powerful π-electron acceptor, which forms charge-transfer complexes even with weak π-donors, such as naphthalene, phenanthrene, and triphenylene [2]. Probably due to its facile hydrolysis and, thus, inconvenient handling, however, MTA has so far received little attention. We report here the crystal structures of MTA and of its 1:1 molecular complexes with triphenylene and 9,10-dimethylanthracene, *i.e.*, with a relatively weak and a relatively strong π-donor, respectively. The work was primarily undertaken to address the following questions: *i*) Is the MTA molecule planar with full D_{3h} symmetry despite the unfavorable directionality of the carbonyl lone pairs and possible ensuing non-bonding O $\cdots$ O repulsions? *ii*) Is the molecular packing of MTA in the solid state dominated by edge-to-face or face-to-face contacts, and how does it relate to the packing of other comparable hexasubstituted benzenes? *iii*) Are the bond lengths of the benzene ring of MTA equal despite the adjacent bond-angle distortions imposed by the fused five-membered anhydride rings?

Scheme 1. Kekulé *Resonance Forms of MTA*



Experimental. – *Crystal-Structure Analyses*1)*.* MTA was prepared by dehydrating mellitic acid (benzenehexacarboxylic acid) with AcCl [2a]. The crystallographic X-ray measurements were performed on a *Nonius* four-circle diffractometer equipped with a *CCD* area counter using Mo radiation ($\lambda = 0.71069$ Å). The structures were solved and refined with the programmes SHELXS97 and SHELXL97, resp. In the following, $R1$ values include reflections with $F > 4\sigma(F)$, $wR2$ values all observed reflections.

Mellitic Trianhydride (*MTA*). Octahedrally shaped crystals were obtained from acetone by slowly removing the solvent with P_2O_5 in a desiccator; they were sealed in glass capillaries under dry conditions. Cubic crystal system, space group *Pa*3 (No. 205), $Z = 4$, $a = 10.1384(5)$ Å; $T = 150$ K; $d_x = 1.836$ g cm^{-3} (densities at r.t.: $d_x = 1.818$, $d_m = 1.81$ g cm^{-3}); $R1 = 0.039$ (549 reflections). $wR2 = 0.111$ (602 reflections); $\Delta\varrho_{\text{res}}$ (max) = 0.41 eÅ^{-3}. We have evidence that, around 145 K, MTA undergoes a phase transition. According to X-ray measurements below this temp., the crystal symmetry is probably orthorhombic, but no detailed structural information could be obtained.

1:1 Molecular Complex of MTA and Triphenylene. Orange-red monoclinic needles from acetone; space group *Pc* (No. 7), $Z = 2$, $a = 7.0803(2)$, $b = 9.1118(4)$, $c = 16.6690(7)$ Å, $\beta = 98.657(2)°$; $T = 100$ K; $d_x = 1.613$ g cm^{-3} (at r.t.: $d_x = 1.576$, $d_m = 1.59$ g cm^{-3}); $R1 = 0.033$ (2292 reflections), $wR2 = 0.085$ (2425 reflections); $\Delta\varrho_{\text{res}}$ (max) = 0.29 eÅ^{-3}.

1:1 Molecular Complex of MTA and 9,10-Dimethylanthracene. Bluish-violet monoclinic needles from hot butan-2-one through slow cooling; space group *Cm* (No. 8), $Z = 2$ (molecular mirror symmetries), $a = 9.9882(6)$, $b = 14.3579(8)$, $c = 7.0973(3)$ Å, $\beta = 95.867(3)°$; $T = 150$ K; $d_x = 1.622$ g cm^{-3} (at r.t.: $d_x = 1.581$, $d_m = 1.59$ g cm^{-3}); $R1 = 0.041$ (1041 reflections), $wR2 = 0.108$ (1100 reflections); $\Delta\varrho_{\text{res}}$ (max) = 0.31 eÅ^{-3}.

Results and Discussion. – Space group and unit-cell content of MTA (*Pa*3, $Z = 2$) require molecular S_6 symmetry, which cannot be satisfied within an ordered structural model. Accordingly, the MTA molecules are seen to be orientationally disordered in the crystal with the two orientations related by an inversion center or an S_6 operation (*Fig. 1,a*). Except for the six-membered-ring C-atoms, both orientations are well resolved without applying constraints in the structure refinement. The MTA molecules

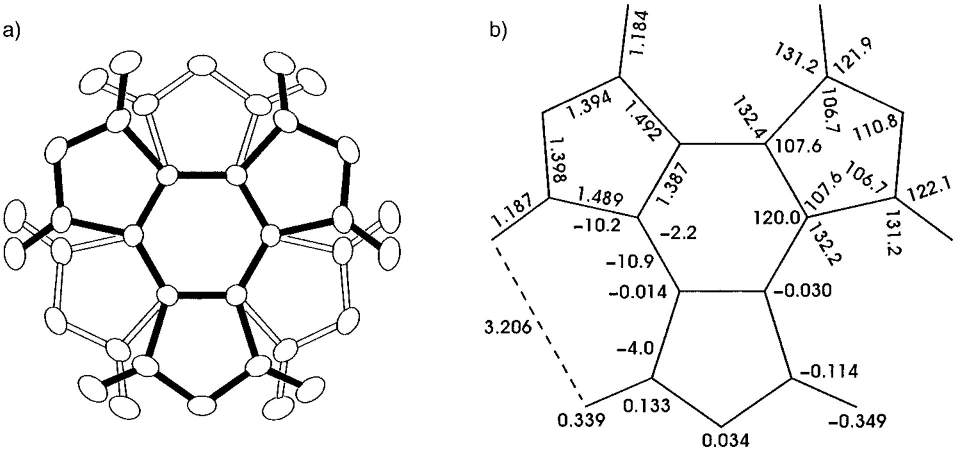


Fig. 1. a) *Orientational disorder of MTA* (with vibrational 50% ellipsoids). b) *Structural data of MTA* (crystallographic C_3 symmetry; bond lengths [Å], bond angles and torsion angles [°], distances from molecular best plane [Å]; estimated average standard deviations: 0.002 Å, 0.2°)

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1) Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the *Cambridge Crystallographic Data Centre* as deposition No. CCDC-135999–CCDC-136001. Copies of the data can be obtained, free of charge, on application to the CCDC, 12 Union Road, Cambridge CB21EZUK (fax: +44(1223)336033; e-mail: deposit@ccdc.cam.ac.uk).

corresponding to a particular orientation have C_3 symmetry and approximate D_3 symmetry.

The MTA molecules deviate significantly from D_{3h} symmetry and are distinctly non-planar in the crystal taking the chiral shape of a shallow three-bladed propeller, in accord with symmetry D_3 (*Figs. 1* and *3*). The structures of both orientations residing on a particular crystal site have opposite chirality, as dictated by its crystallographic average symmetry S_6. The non-planar distortions may be quantified by various geometrical parameters; for example, by the dihedral angle between the mean planes of the benzene and anhydride rings, which adopts the value of 4.9°, or by the individual atoms' distances from the molecular best plane (*Fig. 1,b*). As to the origin of the non-planar distortions, intramolecular non-bonding repulsions between the carbonyl O-atoms may be rather safely ruled out: on the one hand, the measured corresponding $O \cdots O$ distance is 3.206(2) Å, *ca.* 0.4 Å longer than the *van-der-Waals* sum of radii, and, on the other hand, the $O{=}C{-}C$ angles are practically the same as in pyromellitic [3a], phthalic [3b], and maleic anhydride [3c] (symmetry-averaged values 131.2 and 130.9, 131.8, 131.6°, resp.). If there were any sizable non-bonding repulsions between the carbonyl O-atoms in MTA, one would have to expect larger $O{=}C{-}C$ angles as compared to the other anhydrides irrespective of their already widened values. This is so since $O{=}C{-}C$ angle opening is a geometrically much more direct mechanism of increasing the $O \cdots O$ separations than through non-planar molecular distortions. The latter lead to an increase of the $O \cdots O$ separations of less than 0.1 Å (compare the average $O \cdots O$ distances and $O{=}C{-}C$ angles of 3.125 Å and 131.6°, resp., applying to the planar MTA molecules engaged in the two molecular complexes reported further below; *cf. Fig. 4,b*). Therefore, intermolecular packing effects are probably responsible for the non-planarity of MTA in the solid state, as detailed in the following.

The very highly symmetric packing pattern of MTA is shown in *Fig. 2,a*, and it may be seen that every MTA molecule is surrounded by 12 neighbor molecules after the manner of cubic closest packing. The crystal architecture of MTA is remarkable in that the intermolecular contacts are exclusively of the edge-to-face type, although we are dealing with a probably planar (in the gas phase) molecule and π-system of substantial size. In particular, short intermolecular contacts between the five-membered-ring O-atoms and the carbonyl C-atoms of 2.880(2) and 2.888(2) Å (depending on which of the two disordered molecular orientations is involved; *cf. Fig. 3*) may be identified, which are probably attractive, since they correspond to a nucleophile-electrophile type of interaction. This interaction is improved by some pyramidalization of the electrophilic carbonyl C-atom towards the nucleophilic five-membered-ring O-atom, and by some opening of the $O \cdots C{=}O$ angle beyond 90° in the sense of some progress along the reaction coordinate of the nucleophilic attack of the five-membered-ring O-atom on to the carbonyl C-atom (*Scheme 2*) [4]. Now, as an inspection of *Fig. 3* shows, each MTA molecule in the crystal has grouped 3 neighbor molecules each above and below the molecular plane in octahedral fashion, with their five-membered-ring O-atoms pointing towards the carbonyl C-atoms of the central molecule. Thus, it is readily seen that the above distortions supporting the nucleophile-electrophile interaction $\rangle O \cdots C{=}O$ (*Scheme 2*) quite naturally lead to the non-planar, propeller-like shape of MTA in the crystal. The observed pyramidal displacements of the carbonyl C-atoms (referred to the plane of the 3 bound atoms) amount to 0.015(2) and 0.033(2) Å, and

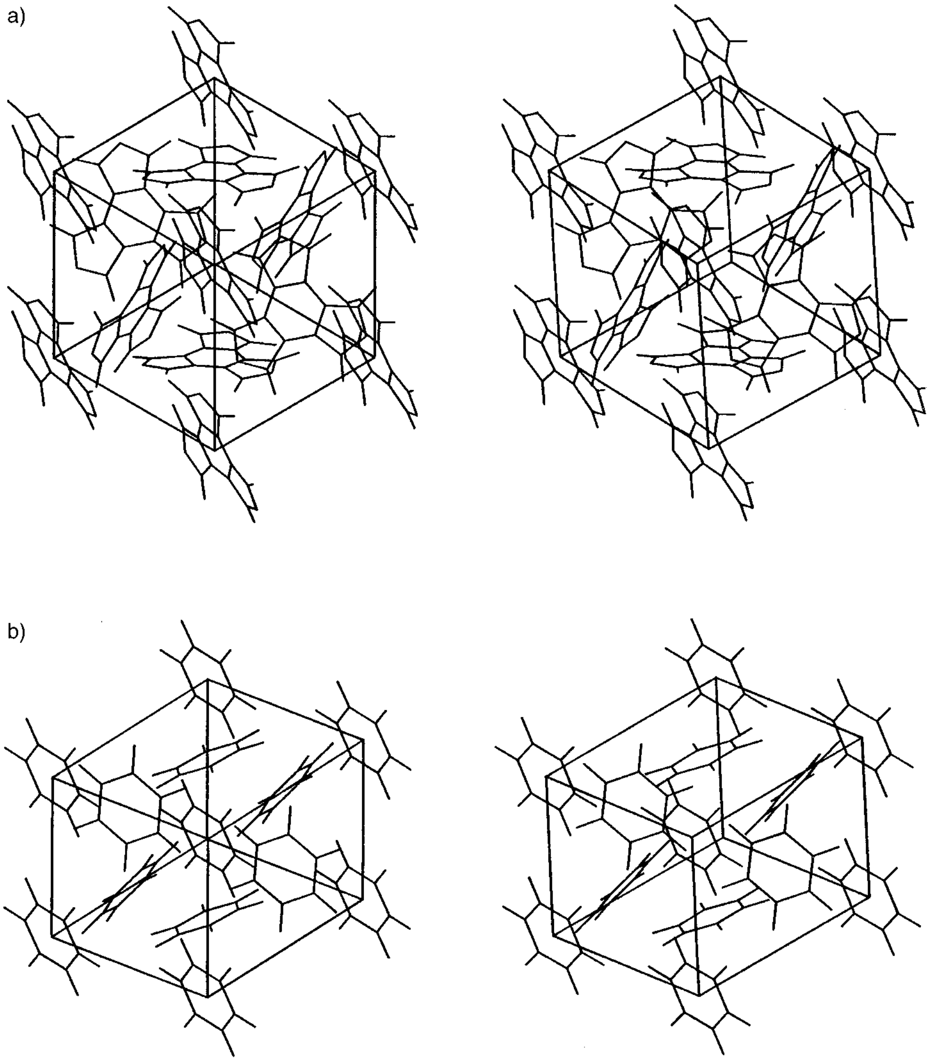


Fig. 2. a) *Stereoview of the crystal structure of MTA along the body diagonal of the cubic unit cell* (for left-hand member of stereo pair). The orientational disorder is not evident from the drawing; the alternative molecular orientations have been omitted in such a way that the architecture shown corresponds to the fictitious space group $P2_13$. b) *Corresponding stereoview of the related crystal structure of benzene, based on the atomic coordinates of* [11] (see text for details of comparison).

indeed occur towards nucleophilically interacting five-membered-ring O-atoms of neighbor molecules. The observed values of the O···C=O angles are 96.7(2) and 90.4(2)°, respectively, depending on which of the disordered molecular orientations is involved, and would be reduced to 86.5 and 80.3°, respectively, if the MTA molecules were planar. These structural distortions ascribed to the nucleophile-electrophile interactions $>$O···O···C=O are well within the range typically recorded previously [4].

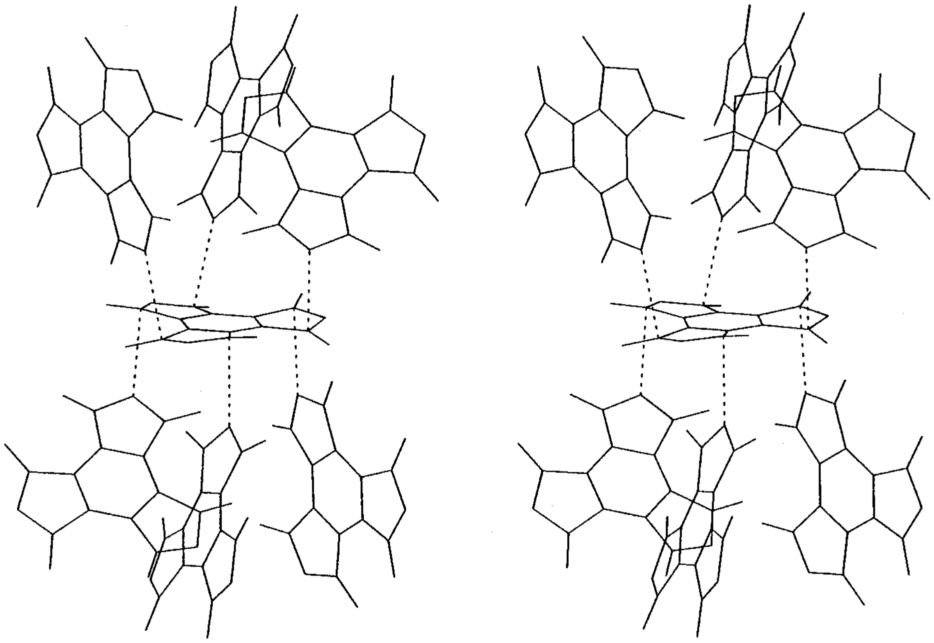
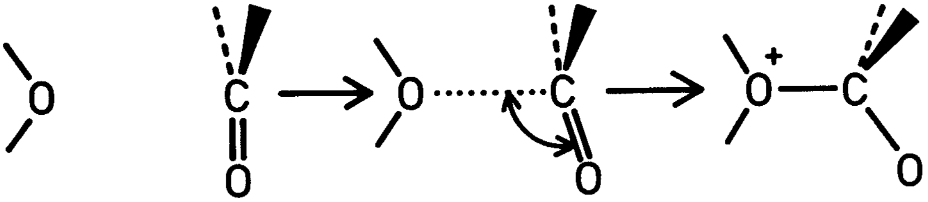


Fig. 3. *Stereoview of a MTA molecule surrounded by 6 neighbor molecules in octahedral disposition to illustrate the nucleophile-electrophile interactions responsible for the non-planar, propeller-like molecular distortions* (>O···C=O interactions dashed; average corresponding non-bonding distance, 2.88 Å). Only one of the two disordered molecular orientations is drawn; note that their statistical weight is thus only 50%.

Scheme 2. *Intermolecular Nucleophile-Electrophile Interaction between Five-Membered-Ring O-Atoms and Carbonyl C-Atoms in Crystalline MTA, Seen as a Transient Configuration of a Nucleophilic Addition Reaction* [4] (O···C=O angle indicated)



(Note that, due to the stoichiometry of MTA and the orientational disorder, on average every MTA molecule in the crystal is surrounded octahedrally by only 3 neighbor molecules in the proper orientation as to provide the discussed nucleophile-electrophile approach (*Fig. 3*).

Summarizing the discussion at this point, we conclude that, in the gas phase, the MTA molecule is planar with D_{3h} symmetry yet rather flexible such that crystal-packing forces suffice to effect substantial non-planar distortions, in particular towards a chiral propeller-shaped structure of D_3 symmetry. The fact that MTA is planar in its molecular complexes with triphenylene and 9,10-dimethylanthracene (see below) corroborates this view. Good-quality vibrational calculations of MTA should be instructive, and they are facilitated by the high molecular symmetry. A very low-lying A_1'' vibrational mode is expected, the normal coordinate of which corresponds to a path, along which two equivalent non-planar propeller conformations of opposite chirality interconvert (*via* the achiral D_{3h} minimum) under conservation of D_3 symmetry.

The unusual crystal-packing characteristics of MTA call for comparisons, and one wonders whether they are unique. A fitting case in place is benzene hexacarbonitrile $C_6(CN)_6$, again a derivative of mellitic acid and a very strong π-acceptor. The crystal structure of this compound is known (at 120 K) [5], and it indeed turns out to be strikingly similar to that of MTA. The space group of benzenehexacarbonitrile is *Pa*3, too, with 4 molecules of symmetry S_6 in the unit cell, and the intermolecular contacts are again exclusively of the edge-to-face type, much like for MTA. However, within experimental error, the hexacarbonitrile molecules are planar in the solid state, and this may be understood through the observation that the nucleophilic cyano N-atoms are not sitting above the electrophilic cyano C-atoms of neighbor molecules but rather on top of the midpoint of lines interconnecting the C-atoms of *ortho*-positioned CN groups. Not surprisingly, the hexacarbonitrile molecules are ordered in the crystal, due to their higher sixfold symmetry D_{6h}. A density of only 1.21 g cm^{-3} is reported for benzenehexacarbonitrile (X-ray value; no macroscopic density measured) [5]. For an organic compound without hydrogen, this value is very low indeed. The likewise hydrogen-free MTA crystals have the high density of 1.82 g cm^{-3} (see *Experimental*), much better in accord with expectation. It follows that the crystal lattice of the hexacarbonitrile involves relatively large cavities [5] (around S_6 sites), which might possibly be occupied by disordered molecules of acetonitrile, from which the crystals were grown.

Benzenehexamine $C_6(NH_2)_6$, a very strong π-donor [6], also crystallizes in space group *Pa*3 with 4 molecules in the unit cell, and the molecules are again packed in edge-to-face fashion, very similar and isomorphous to MTA and benzenehexacarbonitrile. This time the molecules are held together by weak H-bonding, with the N lone pairs and the electron-rich benzene π-system functioning as H acceptors.

Hexaethinylbenzene $C_6(C{\equiv}CH)_6$ [7], could be another candidate adopting the cubic edge-to-face packing pattern of MTA, benzenehexacarbonitrile, and benzenehexamine, possibly with the help of stabilizing guest molecules. T-Shaped H-bonds with the triple-bond π-systems as H acceptors could feature prominently here as observed in acetylene itself. Two forms of acetylene crystals have been characterized [8]; interestingly, the high-temperature form has again space group *Pa*3 with 4 molecules in the unit cell, and the packing pattern is related to that of our three cases discussed above. The low-temperature crystal form of acetylene is orthorhombic, and is characterized by extensive T-shaped H-bonding as suggested for hexaethinylbenzene. Unfortunately, crystallographic measurements of hexaethinylbenzene would appear to be hampered by the compound's limited chemical stability and solubility [7].

The crystal packing of hexafluorobenzene [9a] is edge-to-face dominated, but otherwise not related to MTA. The crystal structures of hexafluoro- [9a], hexachloro- [9b], hexabromo- [9c], and hexaiodobenzene [9d] are all isomorphous with herringbone patterns of molecular stacks involving face-to-face contacts rather different from the crystal architectures of MTA, benzenehexacarbonitrile, and benzenehexamine. This holds similarly for hexamethylbenzene [9e]. However, all-*trans*-hexachlorocyclohexane $(CHCl)_6$, with all Cl-atoms positioned equatorially, crystallizes in space group *Pa*3 with 4 molecules in the unit cell [10], and the crystal packing is again strikingly similar to that of the above three isomorphous hexasubstituted benzenes with this cubic space group. In this fourth case, non-bonding $C-H\cdots Cl$ interactions appear to play a role in establishing the intermolecular edge-to-face contacts.

Interestingly, the crystal structure of benzene itself is also related to that of MTA, benzenehexacarbonitrile, and benzenehexamine. Benzene crystallizes in the orthorhombic space group *Pbca* with 4 molecules in the unit cell (molecular centrosymmetry) [11], and the intermolecular contacts are of the edge-to-face type, *i.e.*, weak H-bonds with the π-electron systems functioning as H acceptors.

Because of the central importance of benzene, featuring the prototypal aromatic molecule, a packing stereo diagram is reproduced in *Fig. 2,b*, for comparison with the crystal structure of MTA (*Fig. 2,a*). The correspondence of both molecular arrays is evident, and becomes still clearer by noting that space group *Pbca* is a subgroup of *Pa*3. The spatial disposition of the C_i and S_6 sites, respectively, in both space groups is the same and corresponds to (distorted) cubic closest packing. The crystal structure of benzene is thus a distorted variant of that of MTA, which is rather unparalleled in the realm of mono- and polycyclic aromatic hydrocarbons. It is intriguing to see that the unique chemical and structural properties of the benzene molecule are matched by an equally singular crystal structure, which, however, does not attain the highest possible symmetry, as realized in the crystals of MTA, benzenehexacarbonitrile, and benzenehexamine[2]).

The C,C bond lengths of the benzene ring of MTA are of interest, since the two non-equivalent bonds, endocyclic and exocyclic with respect to the five-membered rings, could, in principle, adopt different lengths due to the substantially different bond-angle distortions experienced by these bonds. These angle distortions are obviously a consequence of the fact that the angles in a planar five-membered ring are on average 12° smaller than 120°. The exocyclic inner C,C bond flanks two widened bond angles of 132.4°, whereas the endocyclic inner C,C bond flanks two compressed bond angles of 107.6 (average values; *Fig. 1,b*). Using a simple empirical force-field model, one would expect the exocyclic C,C bond to be shortened and the endocyclic C,C bond to be stretched with respect to a common (unstrained) reference length, because bilinear force-field cross-terms between bond lengths and adjacent bond angles usually have a positive sign [12]. Simple hybridization considerations would lead to this conclusion, too. More recent and more elaborate theoretical investigations, however, indicated that, in systems similar to MTA, no bond-length alternation should show up in the central benzene ring, *i.e.*, no so-called '*Mills-Nixon* effect' should be operative [13].

Evidently, as a consequence of the 50:50 orientational disorder, the problem of central bond-length alternation is completely obscured in the crystal structure of MTA. The two superimposed molecular orientations together constitute (average) S_6 symmetry such that both non-equivalent inner C,C bond lengths become equalized, and no resolution is possible. 1:1 Charge-transfer complexes of MTA with the weak π-

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[2]) **Added in proof:** The approximate *Pa*3 symmetry of the crystal structure of benzene has been noted previously: J. D. Dunitz, in 'The Crystal as a Supramolecular Entity', Ed. G. R. Desiraju, John Wiley, Chichester, 1996, p. 1.

donor triphenylene and the strong π-donor 9,10-dimethylanthracene were, therefore, crystallized and their crystal structures determined (see *Experimental*). In both cases, no disorder phenomena are perceptible, such that meaningful conclusions as to the bond-length problem at hand may be drawn. Subtle disorder in terms of a superposition of the two *Kekulé* forms of MTA (*Scheme 1*) is possible in principle, yet extremely unlikely, since both forms are not equivalent and are almost certainly defined within a one-minimum potential (compare the more serious case of benzene with two equivalent *Kekulé* forms [14]). The influence of the π-donors on the in-plane MTA geometry should be negligibly small, given the abundantly available X-ray evidence on numerous other organic charge-transfer π-complexes.

The crystal structures of the two molecular complexes of MTA studied are non-centrosymmetric and polar (space groups *Pc* and *Cm*, resp.; see *Experimental*), which, for donor-acceptor systems, is obviously of interest, in particular with regard to non-linear optical properties. The solid-state architectures of the complexes are characterized by the usual face-to-face stacks of alternating donor and acceptor molecules. The molecular planes in the stacks deviate from orthogonality with respect to the stacking axis in both complexes by practically the same angle, 22°. The donor-acceptor separations in the stacks are on average 3.30 and 3.28 Å, respectively, for MTA · triphenylene and MTA · 9,10-dimethylanthracene. *Fig. 4* offers donor-acceptor overlap diagrams, as well as relevant structural data, of MTA in the two complexes. There are two non-equivalent overlap possibilities in each donor-acceptor stack of both complexes (*Fig. 4,a*, top and bottom). The largest C skeleton triphenylene and 9,10-dimethylanthracene have in common corresponds to a 1,2,3,4-tetrasubstituted naphthalene system, and it may be seen from *Fig. 4,a* (top) that, in one of the two possibilities, MTA overlaps with this common subunit of both π-donors in a similar mode. Though still perceptible, in the second overlap possibility (*Fig. 4,a*, bottom) this correspondence is much more crude and debatable.

The MTA molecule and the donors are essentially planar in both complexes with D_{3h} symmetry well approximated for MTA. The inner six-membered-ring bond lengths of MTA in the two complexes (*Fig. 4,b*) agree within experimental error, and no localization effects are perceptible. There is thus no experimental basis for any discussion in terms of the '*Mills-Nixon* effect' [13] in MTA. As expected, a comparison of the geometry data given in *Fig. 1,b*, and *Fig. 4,b*, furnishes no significant differences among the 3 independent in-plane MTA geometries measured. It is finally noted that the anisotropic temperature motion of MTA in the three structures reported may be well accounted for by a rigid-body model from which a characteristic librational motion around the 3-fold axis emerges. This may already be gleaned qualitatively from the shape of the vibrational ellipsoids of *Fig. 1,a*, and *Fig. 4,a*.

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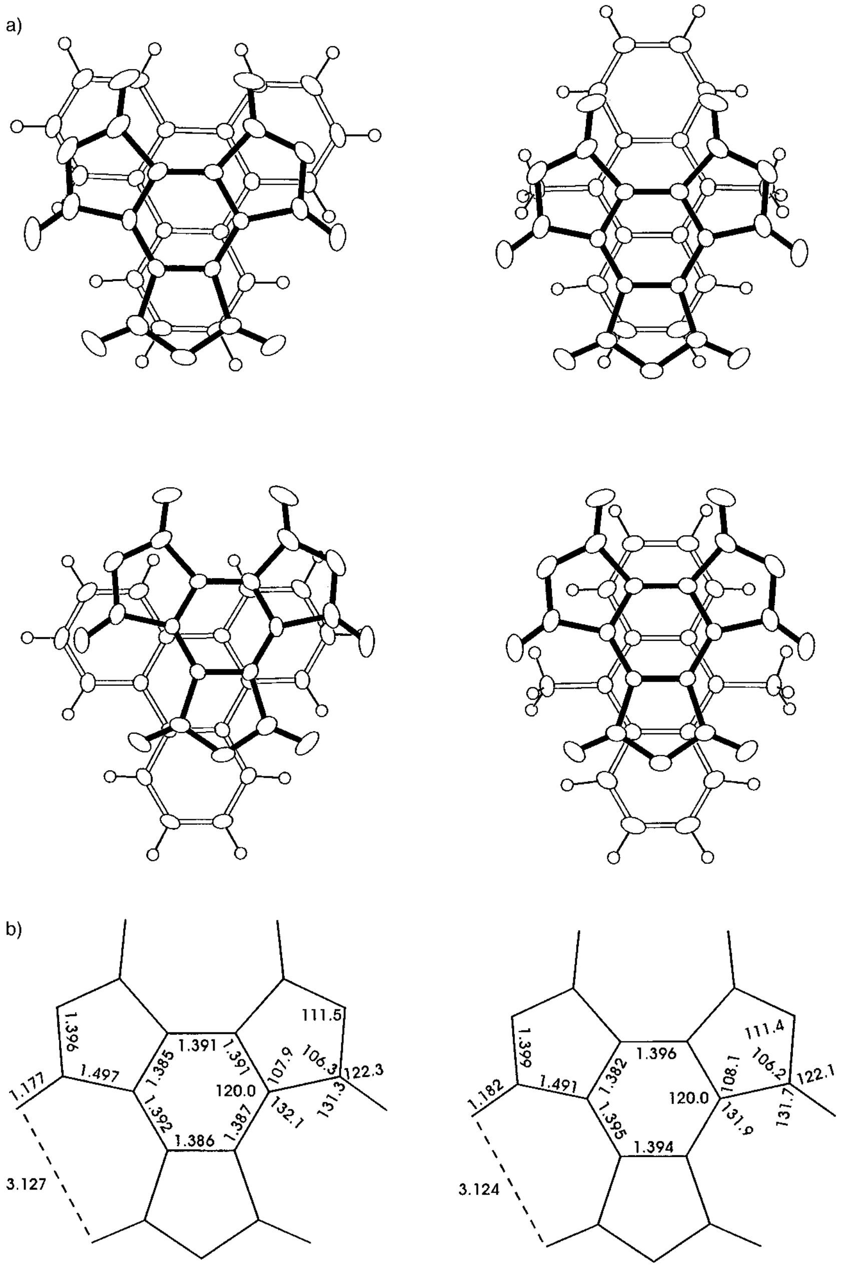

Fig. 4. a) *Donor-acceptor overlap diagrams of the 1:1 molecular charge-transfer complexes of MTA with triphenylene and 9,10-dimethylanthracene* (two non-equivalent overlap possibilities in both complexes: top and bottom; 50% ellipsoids; average separations of molecular planes, 3.30 and 3.28 Å, resp.). b) *Geometry data of MTA in these complexes, averaged over D_{3h} symmetry, except for the* C,C *bond lengths of the benzene ring, whose individual, crystallographically independent values are given* (representation as in *Fig. 1,b*; estimated average standard deviations: 0.003 Å, 0.3°).

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