the calculated mean plane and σ = standard deviation of Δ] are 27·7 (six atoms) and 38·5 (six atoms), respectively. A dihedral angle of 159·9 (1)° between these planes is observed. The *o*-dimethoxy groups are coplanar with the benzene ring as usual (Caillet, 1982; Karle, Flippen-Anderson, Chiang & Lowrey, 1984). The arrangement of the three adjacent methoxy groups is typical (Miravitlles, Solans, Bladé-Font, Germain & Declercq, 1982; Rius, Molins, Miravitlles & Bladé-Font, 1984; Vijayalakshmi, Rajan & Srinivasan, 1987). Thus, the C(13)—O(24)—C(25) group is nearly coplanar with the benzene ring, while the other groups, C(11)—O(20)—C(21) and C(12)—O(22)—C(23), point outside the main rings as can be deduced from Table 3. The C(27)—O(28) and C(27)—N(26) bond distances are 1·227 (3) and 1·396 (3) Å, respectively and these values compare well with those reported for other secondary amides. The observed lengthening of the C—O bond and the shortening of the C—N bond, together with the planarity of the C(9)—N(26)—C(27)—O(28) fragment (see Table 3) are characteristic of an amide function and demonstrate π delocalization. The torsion angles reported in Table 3 describe the conformation of the molecule and show that there is a favoured rotamer with the alkyl substituent at the N atom in a *cis* disposition relative to the carbonyl O atom. Steric hindrance from the different chemical groups is minimized by adoption of a staggered conformation at the central C(8)—C(9) bond, with aryl groups in an *anti* disposition.

The Bischler–Napieralski cyclization of the title compound (assuming that qualitative comparisons can be made concerning the conformation of the molecule in the liquid and solid states) requires that the trimethoxyphenyl ring approaches the carbon attack of the formyl group, which is turned out away from this ring. The disposition of the formyl group could explain the failure of the Bischler–Napieralski reaction to form isoindoles.

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Investigation of the Geometry of Substituted Dinitroanilines. III. Structural Study of *N*,*N*-Dicyclohexyl-2,4-dinitroaniline*

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Abstract. $C_{18}H_{25}N_3O_4$, triclinic, $P\bar{1}$, M_r = 347·4, a = 9·881 (1), b = 12·687 (2), c = 15·438 (5) Å, α = 109·65 (1), β = 90·94 (2), γ = 95·78 (1)°, V = 1811 (1) Å^3, Z = 4, $F(000)$ = 744, D_x = 1·27 g cm^{-3}, T = 296 (2) K, Mo $K\alpha$ radiation, λ = 0·71073 Å, μ(Mo $K\alpha$) = 0·89 cm^{-1}, final R = 0·047, wR = 0·052 for 2294 observed independent reflections. The two crystallographically non-equivalent molecules have similar valence geometry, but different conformation. Both are non-planar with their amino and *o*-nitro groups twisted out of the mean plane of aromatic rings. The four crystallographically non-equivalent cyclohexane rings have chair conforma-

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tions. Non-additivity of the substituent effects on the benzene ring geometry is observed.

Introduction. Substituted 2,4-dinitroanilines and other conjugated organic molecules display large hyperpolarizabilities (for a recent review see Zyss & Tsoucaris, 1990). However, misorientation of the molecules in the solid or the existence of inversion symmetry may decrease or annul the non-linear optical macroscopic properties. In order to gain insight into the factors that influence the macroscopic behavior of the systems mentioned we have conducted the study of the title compound as part of an investigation of 2,4-dinitroanilines with bulky substituents (Punte, Rivero, Socolovsky & Nudelman, 1987, 1989, 1991).

Experimental. Crystals of the title compound, synthesized according to a procedure similar to that described by Brady & Cropper (1950), and crystallized by slow evaporation from methanol, were kindly provided by S. Socolovsky and N. Nudelman. A yellowish irregular-shaped crystal of approximate dimensions 0·3 × 0·25 × 0·35 mm was used for data collection. The measurements were performed on an automatic four-circle Huber diffractometer with graphite-monochromated Mo *Kα* radiation. Lattice parameters were obtained from the setting angles of 20 reflections in the range $18{\cdot}5 < 2\theta < 34°$. Data were collected in the ω step scan mode, scan width 1°, min.–max. scan speed 0·77–4·62° $\min^{-1}$, to $2\theta_{\max} = 40°$; index range h: $-9,9$; k: $-12,11$; l: 0,14. Three standard reflections ($0\bar{1}\bar{2}$, $2\bar{2}0$, 420) measured every 90 min showed 4% intensity variation. Lorentz and polarization corrections were applied. No absorption correction. From 3361 measured independent intensities, 2294 were considered observed on the criterion $I > 3\sigma(I)$, where $\sigma(I)$ is the standard deviation based on counting statistics. The structure was solved using direct methods (*MULTAN*80; Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1980). The solution showed two molecules per asymmetric unit. The structure was refined by block-matrix least squares (*SHELX*76; Sheldrick, 1976), one block for each molecule, based on F's. After an anisotropic refinement of the thermal parameters the H atoms were stereochemically positioned. Further refinements were performed with the H atoms riding on their bound C atoms; overall temperature factors were refined for H atoms of the methyl groups, for H atoms of the cyclohexyl ring and for H atoms of the benzene ring. Isotropic secondary-extinction correction of the form $F' = F(1 - CF^2/\sin\theta)$ was applied on F_c, refined C value: $4{\cdot}76 \times 10^{-3}$. The final refinement, with 466 parameters, converged to $R = 0{\cdot}047$, $wR = 0{\cdot}052$; weighing scheme: $w = 1{\cdot}258 \times [\sigma^2(F_o) + 0{\cdot}0012F_o^2]$, maximum shift/e.s.d. $= 2 \times 10^{-2}$. Minimum and maximum values of the residual electron density were $-0{\cdot}18$ and 0·14 e Å^{-3}, respectively. Scattering factors for H atoms, scattering factors and anomalous-dispersion corrections for O, C and N atoms were provided by *SHELX*76. Calculations were carried out on a MicroVAXII system using *SHELX*76, *MULTAN*80, *PLUTO* (Motherwell & Clegg, 1978), *ORTEP* (Johnson, 1965) and *THMA*11 (Maverick & Trueblood, 1988).

Fractional atomic coordinates and equivalent isotropic thermal parameters for the non-H atoms are presented in Table 1, and bond lengths and angles within the molecule and selected intermolecular contacts are given in Table 2.* The numbering scheme for both molecules is shown in Fig. 1.

Discussion. Least-squares calculations show that the aromatic rings in the two non-equivalent molecules are non-planar. Molecule *A* presents a boat-like deformation; C1*A* and C4*A* distances from the mean plane defined by the other ring carbons are 0·088 (4) and 0·032 (5) Å, respectively. In molecule *B* atoms C1*B* and C5*B* are out of the plane defined by the remaining ring atoms; their distances to the plane are −0·116 (4) and 0·042 (4) Å, respectively. In both molecules the aromatic ring shows deviations from the unsubstituted benzene geometry. Departures of the endocyclic angles from the ideal sp^2 value are similar in both molecules.

Domenicano & Murray Rust (1979) (DMR) and Norrestam & Schepper (1981) (NS) angular parameters were used to estimate the non-additivity of the substituent effects on the angular geometry of the ring. Dimethylamino-group parameters replaced the unavailable dicyclohexylamino-group parameters. Experimental and predicted values are shown in Table 2. The non-additivity parameter (NAP) calculated after the DMR model is 6·7° in *A* and 7·5° in *B*. NAP is defined as $\mathrm{NAP} = \sum|\theta_i - \theta_i(\mathrm{calc})|$ (Krygowsky, 1990, and references therein). The inclusion of the NS parameter for highly twisted NO_2 groups for the *o*-NO_2 substituent and with DMR parameters for the other substituents gives an even greater non-additivity, NAP = 9·6° in *A*, and 8·9° in *B*. NAP values obtained using both models are larger than $3\sigma(= 1°$ in *A* and *B*), thus indicating interaction between the substituents.

We found similar figures for endocyclic angles and the NAP parameter in *N*,*N*-diisopropyl-2,4-dinitroaniline (I) (Punte, Rivero, Socolovsky &

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* Lists of structure factors, anisotropic thermal parameters, H-atom parameters, torsional angles and best least-squares planes have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 54059 (28 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Fractional positional parameters and equivalent isotropic thermal parameters* (Å^2) *with e.s.d.'s in parentheses*

$$B_{eq} = (4/3)\sum_i \sum_j \beta_{ij} \mathbf{a}_i . \mathbf{a}_j.$$

	x	y	z	B_{eq}
O1*A*	0·3218 (3)	0·0915 (3)	−0·0454 (2)	6·2 (1)
O1*A*	0·2761 (3)	0·1941 (3)	−0·1250 (2)	7·0 (1)
O3*A*	0·6642 (4)	0·4495 (3)	−0·1335 (3)	7·4 (1)
O4*A*	0·7970 (4)	0·5307 (3)	−0·0124 (2)	7·6 (1)
N1*A*	0·3731 (3)	0·2449 (3)	0·1433 (2)	3·8 (1)
N2*A*	0·3349 (4)	0·1801 (4)	−0·0602 (2)	4·9 (1)
N3*A*	0·6987 (4)	0·4651 (3)	−0·0529 (3)	5·5 (1)
C1*A*	0·4596 (4)	0·2880 (3)	0·0926 (3)	3·8 (1)
C2*A*	0·4340 (4)	0·2724 (3)	−0·0022 (3)	3·8 (1)
C3*A*	0·5086 (5)	0·3302 (4)	−0·0488 (3)	4·1 (1)
C4*A*	0·6202 (5)	0·4020 (4)	−0·0045 (3)	4·2 (1)
C5*A*	0·6598 (5)	0·4132 (4)	0·0846 (3)	4·9 (2)
C6*A*	0·5821 (4)	0·3572 (3)	0·1310 (3)	4·7 (1)
C7*A*	0·4226 (4)	0·2101 (3)	0·2199 (2)	4·0 (1)
C8*A*	0·2233 (4)	0·2433 (3)	0·1292 (3)	3·8 (1)
C9*A*	0·1416 (4)	0·1312 (3)	0·1205 (3)	4·6 (1)
C10*A*	−0·0077 (5)	0·1355 (4)	0·0950 (3)	5·7 (2)
C11*A*	−0·0663 (5)	0·2336 (4)	0·1646 (3)	6·4 (2)
C12*A*	0·0194 (4)	0·3444 (4)	0·1791 (3)	5·6 (2)
C13*A*	0·1684 (4)	0·3397 (3)	0·2035 (3)	4·1 (1)
C14*A*	0·5319 (5)	0·1309 (4)	0·1881 (3)	5·2 (1)
C15*A*	0·5699 (5)	0·0814 (4)	0·2613 (3)	6·4 (2)
C16*A*	0·6094 (5)	0·1736 (4)	0·3528 (3)	6·4 (2)
C17*A*	0·4983 (5)	0·2496 (4)	0·3836 (3)	6·0 (2)
C18*A*	0·4645 (5)	0·3028 (4)	0·3120 (3)	4·9 (1)
O1*B*	0·0409 (4)	0·1062 (3)	−0·5573 (2)	7·5 (1)
O2*B*	0·1396 (4)	0·2222 (3)	−0·6166 (2)	7·6 (1)
O3*B*	−0·2012 (4)	0·4684 (3)	−0·6299 (3)	7·1 (1)
O4*B*	−0·3194 (4)	0·5374 (3)	−0·5121 (2)	8·0 (1)
N1*B*	0·0462 (3)	0·2538 (3)	−0·3579 (2)	4·0 (1)
N2*B*	0·0638 (4)	0·1995 (4)	−0·5622 (3)	5·2 (1)
N3*B*	−0·2325 (4)	0·4788 (3)	−0·5506 (3)	5·6 (1)
C1*B*	−0·0300 (4)	0·2982 (3)	−0·4076 (3)	3·8 (1)
C2*B*	−0·0094 (4)	0·2887 (3)	−0·5011 (3)	3·8 (1)
C3*B*	−0·0700 (4)	0·3496 (4)	−0·5462 (3)	4·2 (1)
C4*B*	−0·1668 (5)	0·4154 (3)	−0·5026 (3)	4·2 (1)
C5*B*	−0·2035 (4)	0·4191 (3)	−0·4160 (3)	4·8 (2)
C6*B*	−0·1386 (4)	0·3618 (3)	−0·3707 (3)	4·4 (1)
C7*B*	−0·0074 (4)	0·2208 (3)	−0·2802 (3)	4·4 (1)
C8*B*	0·1920 (4)	0·2403 (3)	−0·3763 (3)	4·2 (1)
C9*B*	0·2288 (5)	0·1211 (4)	−0·3957 (3)	5·7 (2)
C10*B*	0·3749 (6)	0·1162 (5)	−0·4243 (4)	7·9 (2)
C11*B*	0·4738 (6)	0·2016 (7)	−0·3526 (5)	9·2 (3)
C12*B*	0·4316 (5)	0·3184 (5)	−0·3281 (4)	7·3 (2)
C13*B*	0·2859 (4)	0·3234 (4)	−0·2988 (3)	5·1 (2)
C14*B*	−0·1383 (5)	0·1420 (3)	−0·3091 (3)	5·5 (2)
C15*B*	−0·1734 (6)	0·0906 (4)	−0·2355 (4)	7·6 (2)
C16*B*	−0·1850 (6)	0·1824 (5)	−0·1431 (4)	7·5 (2)
C17*B*	−0·0562 (6)	0·2638 (5)	−0·1151 (3)	6·9 (2)
C18*B*	−0·0186 (5)	0·3148 (4)	−0·1884 (3)	5·4 (2)

Nudelman, 1989) and in *N*-cyclohexyl-*N*-isopropyl-2,4-dinitroaniline (II) (Punte, Rivero, Socolovsky & Nudelman, 1991). In (II) the departure of the endocyclic angles from the predicted values could be explained as being mainly due to hindrance and resonance effects. In the title compound an undoubtedly important *ortho* effect is observed. Indeed, there is extensive rotation of the amino and *o*-nitro groups out of the mean ring planes, the twist angles being $\varphi(\mathrm{N}R_2)_A = 37{\cdot}7\ (2)$, $\varphi(\mathrm{NO_2})_A = 37{\cdot}3\ (2)$, $\varphi(\mathrm{N}R_2)_B = 29{\cdot}8\ (3)$ and $\varphi(\mathrm{NO_2})_B = 46{\cdot}3\ (2)°$, respectively. Besides, N1 and N2 atoms exhibit noticeable deviations, in opposite directions, from the mean phenyl planes. The angles formed by the C1—N1 and C2—N2 bonds with the mean ring planes are 9·9 and 13·0° in *A* and 7·5 and 15·6° in *B*. However, it must be pointed out that in spite of the differences found in the twist angles of the NR_2 and *o*-NO$_2$

Table 2. *Intramolecular bond distances* (Å), *bond angles* (°) *and selected intermolecular distances* (Å) *with e.s.d.'s in parentheses*

Bond distances corrected for librational and torsional effects (Maverick & Trueblood, 1988) in square brackets. Predicted endocyclic bond angles (°) are in the third and fourth columns (Domenicano & Murray-Rust, 1979).

	Molecule *A*		Molecule *B*	
O1—N2	1·215 (7)	[1·234]	1·210 (7)	[1·228]
O2—N2	1·221 (5)	[1·241]	1·220 (6)	[1·236]
O3—N3	1·229 (6)	[1·250]	1·234 (7)	[1·253]
O4—N3	1·222 (5)	[1·243]	1·221 (5)	[1·240]
N1—C1	1·363 (6)	[1·365]	1·353 (6)	[1·357]
N1—C8	1·490 (5)	[1·493]	1·489 (5)	[1·494]
N1—C7	1·486 (5)	[1·491]	1·486 (6)	[1·491]
N2—C2	1·472 (5)	[1·476]	1·471 (6)	[1·477]
N3—C4	1·448 (7)	[1·451]	1·452 (7)	[1·455]
C1—C2	1·424 (7)	[1·427]	1·427 (7)	[1·432]
C1—C6	1·414 (5)	[1·417]	1·419 (5)	[1·423]
C2—C3	1·364 (7)	[1·367]	1·372 (7)	[1·375]
C3—C4	1·366 (6)	[1·370]	1·367 (6)	[1·372]
C4—C5	1·380 (7)	[1·383]	1·378 (7)	[1·382]
C5—C6	1·363 (7)	[1·365]	1·360 (7)	[1·364]
C7—C14	1·524 (6)	[1·528]	1·514 (6)	[1·520]
C7—C18	1·528 (5)	[1·531]	1·529 (5)	[1·536]
C8—C13	1·522 (5)	[1·526]	1·524 (5)	[1·530]
C8—C9	1·526 (5)	[1·529]	1·523 (6)	[1·529]
C10—C9	1·532 (6)	[1·535]	1·518 (8)	[1·523]
C11—C10	1·518 (6)	[1·522]	1·516 (8)	[1·522]
C12—C11	1·512 (7)	[1·514]	1·50 (1)	[1·51]
C13—C12	1·526 (6)	[1·529]	1·517 (6)	[1·522]
C14—C15	1·524 (8)	[1·528]	1·517 (8)	[1·522]
C15—C16	1·515 (5)	[1·519]	1·523 (7)	[1·530]
C16—C17	1·511 (7)	[1·516]	1·513 (8)	[1·519]
C17—C18	1·524 (8)	[1·527]	1·515 (8)	[1·520]

	Molecule *A*	Molecule *B*		
C1—N1—C8	119·6 (4)	120·9 (4)		
C1—N1—C7	122·3 (3)	122·7 (3)		
C8—N1—C7	117·8 (3)	116·4 (3)		
O1—N2—O2	123·0 (4)	123·9 (4)		
O1—N2—C2	118·9 (4)	117·2 (4)		
O2—N2—C2	117·9 (4)	118·8 (5)		
O3—N3—O4	122·7 (4)	122·7 (5)		
O3—N3—C4	118·4 (4)	118·5 (4)		
O4—N3—C4	118·8 (4)	118·8 (4)		
N1—C1—C2	124·0 (3)	124·5 (3)		
N1—C1—C6	122·6 (4)	122·9 (4)		
C2—C1—C6	113·3 (4)	112·7 (4)	116·1 (0)	116·6 (6)*
N2—C2—C1	120·9 (4)	121·9 (4)		
N2—C2—C3	114·7 (4)	113·2 (4)		
C1—C2—C3	123·8 (3)	124·2 (4)	123·8 (6)	125·5 (5)*
C2—C3—C4	118·7 (4)	118·3 (4)	117·6 (5)	116·4 (4)*
N3—C4—C3	119·1 (4)	118·5 (4)		
N3—C4—C5	120·0 (4)	120·8 (4)		
C3—C4—C5	120·8 (5)	120·6 (5)	121·5 (6)	121·2 (6)*
C4—C5—C6	119·6 (4)	120·3 (4)	119·9 (6)	121·1 (5)*
C1—C6—C5	123·0 (4)	123·0 (4)	121·2 (5)	120·9 (4)*
N1—C8—C13	111·5 (3)	111·3 (3)		
N1—C8—C9	114·1 (3)	114·5 (3)		
C13—C8—C9	110·6 (3)	110·2 (4)		
C8—C13—C12	109·6 (3)	108·8 (4)		
C13—C12—C11	111·9 (4)	112·2 (5)		
C12—C11—C10	112·0 (4)	111·3 (6)		
C11—C10—C9	111·6 (3)	112·2 (4)		
C8—C9—C10	109·1 (4)	109·0 (4)		
N1—C7—C14	110·6 (3)	111·4 (3)		
N1—C7—C18	117·5 (3)	117·6 (3)		
C14—C7—C18	111·8 (3)	111·6 (3)		
C7—C14—C15	111·4 (4)	109·8 (4)		
C14—C15—C16	111·0 (4)	110·6 (4)		
C15—C16—C17	111·5 (4)	111·4 (5)		
C16—C17—C18	111·2 (4)	111·8 (4)		
C7—C18—C17	109·4 (4)	109·6 (4)		

O2*A*···C17*B*^{i}	3·478 (7)	O1*B*···C14*B*^{v}	3·377 (5)
O1*A*···C14*A*^{ii}	3·413 (5)	O3*B*···C1*B*^{vi}	3·430 (5)
O3*A*···C13*A*^{iii}	3·488 (6)	O3*B*···C13*B*^{vi}	3·355 (7)
N3*A*···C3*A*^{iii}	3·441 (6)	N3*B*···C2*B*^{vi}	3·462 (5)
C17*A*···O4*B*^{iv}	3·440 (6)	C3*B*···C4*B*^{vi}	3·458 (6)
C18*A*···O4*B*^{iv}	3·497 (5)		

Symmetry codes: (i) x, y, z; (ii) $1-x, -y, -z$; (iii) $1-x, 1-y, -z$; (iv) $-x, 1-y, -z$; (v) $-x, -y, -1-z$; (vi) $-x, 1-y, -1-z$.

* Twist NO$_2$NS parameters for the *o*-nitro substituent.

groups and in the exocyclic angles at C1 and C2 in going from *A* to *B*, the differences between endocyclic angles are within experimental error (see Table 2). This would indicate that either resonance effects or geometrical constraints have a greater influence on the observed NAP values than intermolecular interactions (which however are able to induce the existence of two crystallographically non-equivalent molecules).

In fact, in spite of the *ortho* effect mentioned above the C1—N1 bond lengths [1·363 (6) Å for *A* and 1·353 (6) Å for *B*] indicate some double-bond character. Besides, the overall distribution of bond lengths (corrected for thermal motion and internal torsion of the NO_2 groups) and the coplanarity within experimental error of the *p*-NO_2 group and the mean ring plane [twist angles 3(3) and 1(5)° in *A* and *B*, respectively] indicate a certain degree of quinoid contribution to the resonance of the molecule. On the other hand, intermolecular interactions of different strength are likely, involving O atoms belonging to the *o*-NO_2 group in both molecules, as can be seen in Table 2. Preliminary studies of the geometry of the *N*,*N*-diethyl-2,4-dinitroaniline (III) molecule would indicate that intermolecular interactions might modify the ring geometry more than could be expected from the present findings. Therefore a thorough investigation of (III) and of other substituted 2,4-dinitroanilines is currently under way for disclosing the influence of the different factors mentioned above on the NAP values.

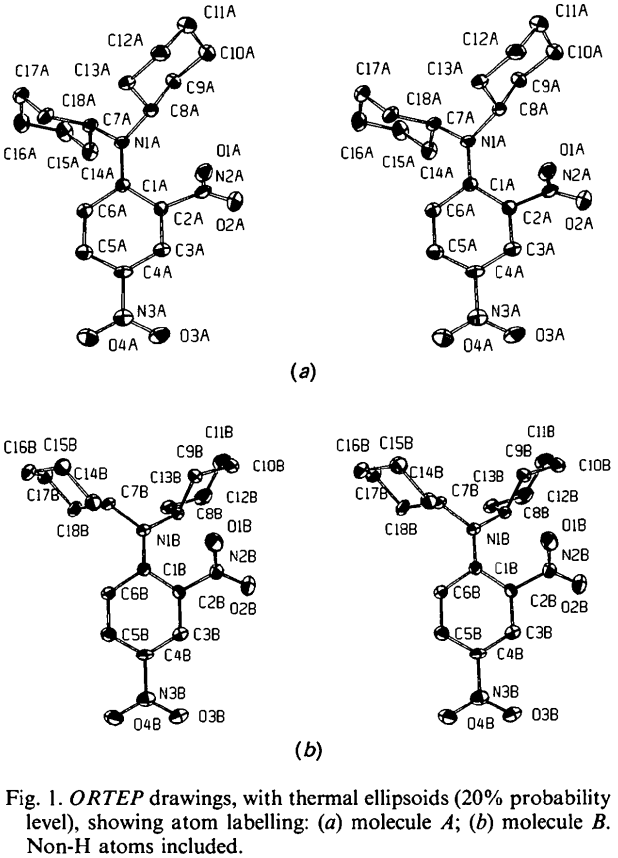

Fig. 1. *ORTEP* drawings, with thermal ellipsoids (20% probability level), showing atom labelling: (*a*) molecule *A*; (*b*) molecule *B*. Non-H atoms included.

The geometry of the NO_2 groups compares well with that found in similar compounds (Holden, Dickinson & Bock, 1972; Maurin & Krygowski, 1987). The four non-equivalent cyclohexane rings are located with their mean planes almost perpendicular to their attached amino group; they have a chair conformation with different deviations from the ideal conformation described by Bucourt & Hainaut (1965). Their mean torsion angles [56·4 (2·6) and 55·8 (1·4)° in *A*; 56·7 (1·1) and 57·0 (2·8)° in *B*] are slightly bigger than the expected ideal value (55°) and the mean endocyclic angles [110·8 (1·1) and 111·0 (0·8)° in *A*; 110·6 (1·4) and 110·8 (0·9)° in *B*] are slightly smaller than the expected ideal value (111·5°).

A *PLUTO* stereoview of the packing in the unit cell is displayed in Fig. 2. It shows that molecules are stacked forming chains along **c**. An antiparallel array of dipolar moments of neighboring chains is observed. This gives rise to the packing of molecules *A* and *B* in non-polar layers centered around $z = 0·0$ and $z = 0·5$, respectively, with the mean phenyl planes of *A* and *B* forming an angle of 77·3 (1)°. This centrosymmetrical packing of the molecules within each layer, probably due to strong intermolecular dipole–dipole interactions, favors a close-packed arrangement of the layers (Kitaigorodskii, 1961) and cancels the macroscopic quadratic electrical susceptibility.

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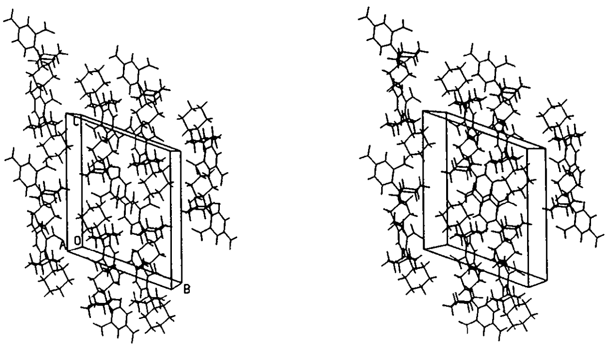

Fig. 2. Stereoscopic view of the molecular packing, **b** across, **a** down.

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Structure of *trans*-2,2,5,5-Tetracyano-3,6-diphenylpiperazine–Propanone (1/2)

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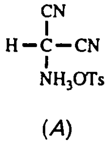
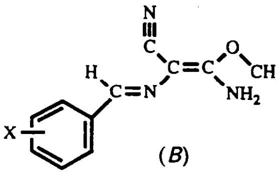
Abstract. *trans*-3,6-Diphenyl-2,2,5,5-piperazinetetracarbonitrile–propanone (1/2), $C_{20}H_{14}N_6.2C_3H_6O$, $M_r = 454{\cdot}50$, monoclinic, $P2_1/n$, $a = 9{\cdot}183\,(2)$, $b = 7{\cdot}3448\,(13)$, $c = 18{\cdot}272\,(3)$ Å, $\beta = 91{\cdot}640\,(15)°$, $V = 1231{\cdot}9\,(4)$ Å^3, $Z = 2$, $D_x = 1{\cdot}23$ Mg m^{-3}, λ(Mo $K\alpha$) $= 0{\cdot}710730$ Å, $\mu = 0{\cdot}075$ mm^{-1}, $F(000) = 480$, $T = 183$ K, final $R = 0{\cdot}061$ for 1591 observed reflections. The structure consists of a piperazine ring in the chair conformation with the phenyl groups occupying equatorial positions. There are two propanone solvent molecules per piperazine unit. All exterior and interior bond angles of the piperazine ring are approximately tetrahedral.

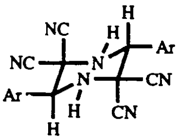
Introduction. Aminopropanedinitrile 4-methylbenzenesulfonate [aminomalononitrile *p*-toluenesulfonate (*A*)] reacts with aromatic aldehydes in methanolic sodium ethanoate to give (*E*,*E*)-4-amino-1-aryl-3-cyano-4-methoxy-2-aza-1,3-butadienes (*B*) and *trans*-2,2,5,5-tetracyano-3,6-diarylpiperazines (*C*) in good to excellent yields (Kim, 1989; Freeman & Kim, 1989, 1991). Infrared, ^{1}H NMR, ^{13}C NMR and ultraviolet spectra, and elemental analyses confirm the empirical formula of the piperazines (*C*) but cannot unambiguously assign the aryl groups to the axial or equatorial positions (diastereomers).



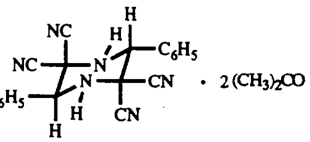
(*A*)



(*B*)



(*C*)



(*D*)

Experimental. Crystals of *trans*-2,2,5,5-tetracyano-3,6-diphenylpiperazine (*D*) were grown by slow evaporation from a solution of the compound in propanone. A colorless crystal of approximate dimensions 0·17 × 0·20 × 0·33 mm was oil mounted on a glass fiber. Data were collected on a Nicolet *P*3 diffractometer equipped with a modified LT-2 apparatus using standard techniques (Churchill, Lashewycz & Rotella, 1977). θ–2θ scans for $2\theta = 4{\cdot}0$–$50{\cdot}0°$

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