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A monoclinic polymorph of 3-amino-4′-(*N*,*N*-diethylamino)-5-methylbiphenyl-2,4-dicarbonitrile

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Key indicators

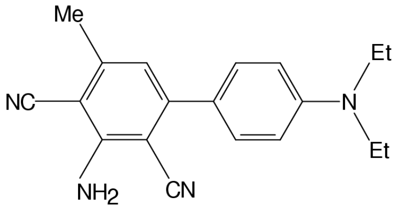
Single-crystal X-ray study
T = 293 K
Mean σ(C–C) = 0.005 Å
R factor = 0.068
wR factor = 0.185
Data-to-parameter ratio = 16.7

For details of how these key indicators were automatically derived from the article, see http://journals.iucr.org/e.

The title compound, $C_{19}H_{20}N_4$, is found to crystallize in both the orthorhombic and the monoclinic crystal systems. In the monoclinic polymorph, the dihedral angle between the two phenyl rings is 53.5 (2)°. Centrosymmetrically related molecules in the crystal are linked by N—H···N hydrogen bonds to form cyclic dimers. The structure of the monoclinic polymorph differs from that of the orthorhombic form reported earlier [Subbiah Pandi *et al.*, (2000). *Cryst. Res. Technol.* **35**, 1373–1381] with regard to the conformation of the biphenyl ring system and the molecular packing.

Comment

Biphenyl derivatives exhibit a wide spectrum of photophysical, biological and laser activities (Shukla *et al.*, 1985; Nieger *et al.*, 1998). We recently reported the structure of 3-amino-4′-(*N*,*N*-diethylamino)-5-methylbiphenyl-2,4-dicarbonitrile, (I), in the orthorhombic system (Subbiah Pandi *et al.*, 2000). Here we report the structural details of a monoclinic polymorph of (I).



(I)

The bond lengths and bond angles observed in the present study agree well with the corresponding values reported for the orthorhombic form. However, the structures of the two polymorphs differ in the conformations of the biphenyl ring system and the diethylamino substituent. The dihedral angle between the two phenyl rings is 53.5 (2)° in the monoclinic form, whereas it is 41.4 (1)° in the orthorhombic form. In the monoclinic polymorph, one of the ethyl groups is twisted 9° more about the C4′—N4 bond compared to that in the orthorhombic form. Even though in both polymorphs the packing of the molecules is stabilized by intermolecular N—H···N hydrogen bonds, the pattern of hydrogen bonding is different. In the monoclinic form, the molecules are linked by N1—H1*A*···N3^{i} hydrogen bonds into cyclic centrosymmetric dimers denoted by the $R_2^2(12)$ (Bernstein *et al.*, 1995) ring system [H1*A*···N3 2.22, N1···N3 3.038 (5) Å and N1—H1*A*···N3 159°; symmetry code (i): 3−*x*, −*y*, −*z*]. In the orthorhombic form, the molecules are linked by N—H···N hydrogen bonds to form zigzag chains.

Experimental

The title compound was synthesized according to the reaction described by Raghukumar *et al.* (2001). The crystals of the monoclinic form were grown at room temperature by slow evaporation of a solution of the compound in ethyl acetate and hexane (1:1). In our earlier work, the crystals of the orthorhombic form were grown from a mixture of ethyl acetate and methanol.

Crystal data

$C_{19}H_{20}N_4$
$M_r = 304.39$
Monoclinic, $P2_1/c$
$a = 9.2925$ (4) Å
$b = 10.8655$ (3) Å
$c = 16.7229$ (5) Å
$\beta = 90.367$ (2)°
$V = 1688.44$ (10) Å^3
$Z = 4$

$D_x = 1.197$ Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 3225 reflections
$\theta = 2.2$–$28.3°$
$\mu = 0.07$ mm^{-1}
$T = 293$ (2) K
Slab, yellow
0.26 × 0.14 × 0.10 mm

Data collection

Siemens SMART CCD area-detector diffractometer
ω scans
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)
$T_{\min} = 0.978$, $T_{\max} = 0.990$
10 442 measured reflections

3496 independent reflections
1313 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.124$
$\theta_{\max} = 26.5°$
$h = -11 \rightarrow 11$
$k = -12 \rightarrow 13$
$l = -20 \rightarrow 20$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.068$
$wR(F^2) = 0.185$
$S = 0.92$
3496 reflections
209 parameters
H-atom parameters constrained

$w = 1/[\sigma^2(F_o^2) + (0.0674P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{\max} < 0.001$
$\Delta\rho_{\max} = 0.19$ e Å^{-3}
$\Delta\rho_{\min} = -0.18$ e Å^{-3}
Extinction correction: *SHELXL*97
Extinction coefficient: 0.010 (2)

Table 1
Selected torsion angles (°).

C12—N4—C4′—C5′	170.9 (3)	C6′—C1′—C1—C6	−52.1 (5)
C10—N4—C4′—C5′	−3.0 (5)	C2′—C1′—C1—C2	−53.5 (5)
C12—N4—C4′—C3′	−10.3 (5)	C4′—N4—C10—C11	83.6 (4)
C10—N4—C4′—C3′	175.8 (3)	C4′—N4—C12—C13	−77.3 (4)

Owing to the poor quality of the crystal, the higher angle reflections were very weak and only 32% of the reflections were found to be observed with $I > 2\sigma(I)$. This resulted in a high $R_{\rm int}$ value. The 2θ maximum was limited to 53° because of the large fraction of weak data at higher angles. All H atoms were fixed geometrically and allowed to ride on the parent non-H atoms.

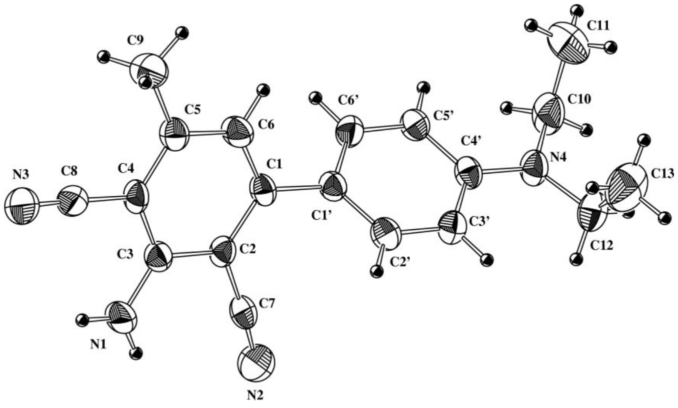


Figure 1
The structure of (I), showing 50% probability displacement ellipsoids and the atom-numbering scheme.

Data collection: *SMART* (Siemens, 1996); cell refinement: *SAINT* (Siemens, 1996); data reduction: *SAINT*; program(s) used to solve structure: *SHELXS*97 (Sheldrick, 1997); program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997); molecular graphics: *ZORTEP* (Zsolnai, 1997); software used to prepare material for publication: *SHELXL*97 and *WinGX* (Farrugia, 1999).

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