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4-{[1-(Methoxycarbonyl)ethylamino](phenyl)-methylidene}-3-methyl-1-phenyl-1*H*-pyrazol-5(4*H*)-one

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Key indicators

Single-crystal X-ray study
T = 293 K
Mean σ(C–C) = 0.007 Å
R factor = 0.042
wR factor = 0.110
Data-to-parameter ratio = 7.3

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

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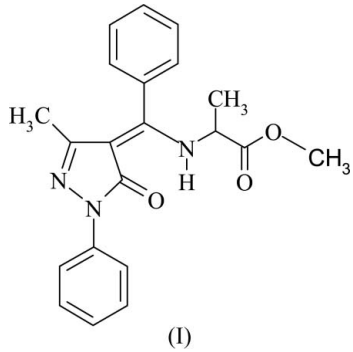
The title compound, $C_{21}H_{21}N_3O_3$, is a neutral potentially tridentate ligand in an enamine-keto form, stabilized by an intramolecular N—H···O hydrogen bond. There are two molecules in the asymmetric unit.

Comment

In recent years, Schiff bases and their metal complexes have been studied widely for their antibacterial activity (Li *et al.*, 1997, 2004). 1-Phenyl-3-methyl-4-benzoylpyrazolon-5-one (PMBP) is widely used and well known for its extractive ability. Both PMBP and its metal complexes also have analgesic activity (Liu *et al.*, 1980; Li *et al.*, 1997; Zhou *et al.*, 1999). Since amino acid esters also possess good antibacterial and biological activities (Xiong *et al.*, 1993), we have studied the reactions of PMBP and amino acid esters.



(I)

A view of the molecular structure of the title compound is shown in Fig. 1. There are two molecules in the asymmetric unit, and the numerical results given here are for one of them; they are not significantly different. Atoms O1, C10, C9, C11 and N3 are coplanar. The dihedral angle between this mean plane and that of the pyrazoline ring is 3.87 (17)°, close to the value of 3.56 (3)° in 4-{[3,4-dihydro-5-methyl-3-oxo-2-phenyl-2*H*-pyrazol-4-ylidene](phenyl)methylamino}-1,5-dimethyl-2-phenyl-1*H*-pyrazol-3(2*H*)-one (Wang *et al.*, 2003). The bond lengths within this part of the molecule (Table 1) lie between classical single- and double-bond lengths, indicating extensive conjugation. The phenyl group bonded to N1 and the pyrazolone ring are approximately coplanar, the dihedral angle between them being 4.73 (17)°; the phenyl group bonded to C11 is perpendicular to the pyrazolone ring, with a dihedral angle of 89.34 (16)°, reducing steric hindrance.

Atoms N3, C18, C20 and O2 of the alanine methyl ester group are not coplanar, the torsion angle being −28.6 (4)°, as seen in ethyl 2-{[(1*Z*)-(3-methyl-5-oxo-1-phenyl-1,5-dihydro-pyrazol-4-ylidene)(phenyl)-methyl]amino}-3-phenylpropan-

oate (Zhang *et al.*, 2005), but different from the situation in some other 4-acylpyrazolone Schiff bases (Zhang *et al.*, 2004; Wang *et al.*, 2003).

A strong intramolecular hydrogen bond is observed (Table 2), stabilizing the enamine-keto form. This is similar to the situation in 4-{[3,4-dihydro-5-methyl-3-oxo-2-phenyl-2*H*-pyrazol-4-ylidene](phenyl)methylamino}-1,5-dimethyl-2-phenyl-1*H*-pyrazol-3(2*H*)-one [N···O = 2.745 (4) Å and N—H···O =146 (4)°; Wang *et al.*, 2003]. In the title compound, atoms O1 and N3 are posible coordinating atoms. Atom O2 is a third possible coordinating atom if there is suitable rotation about the N3—C18 and C18—C20 bonds.

Experimental

The title compound was synthesized by refluxing a mixture of PMBP (15 mmol) and alanine methyl ester (15 mmol) in ethanol (100 ml) over a steam bath for about 4 h. Excess solvent was removed by evaporation and the solution was cooled to room temperature. After four days pale yellow blocks were obtained and dried in air. The product was recrystallized from ethanol, affording pale yellow crystals suitable for X-ray analysis.

Crystal data

$C_{21}H_{21}N_3O_3$
M_r = 363.41
Monoclinic, $P2_1$
a = 8.8118 (18) Å
b = 12.430 (3) Å
c = 18.031 (4) Å
β = 97.749 (2)°
V = 1956.9 (7) Å^3
Z = 4
D_x = 1.234 Mg m^{-3}
Mo $K\alpha$ radiation
Cell parameters from 1621 reflections
θ = 2.3–17.6°
μ = 0.08 mm^{-1}
T = 293.2 (2) K
Block, pale yellow
0.38 × 0.32 × 0.20 mm

Data collection

Bruker APEX2 CCD area detector diffractometer
φ and ω scans
Absorption correction: multi-scan (*SADABS*; Sheldrick, 1996)
T_{min} = 0.934, T_{max} = 0.987
10774 measured reflections
3620 independent reflections
2191 reflections with $I > 2\sigma(I)$
R_{int} = 0.042
θ_{max} = 25.0°
$h = -10 \rightarrow 10$
$k = -14 \rightarrow 14$
$l = -21 \rightarrow 17$

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)]$ = 0.042
$wR(F^2)$ = 0.110
S = 1.06
3620 reflections
494 parameters
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0458P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max}$ = 0.13 e Å^{-3}
$\Delta\rho_{min}$ = −0.15 e Å^{-3}
Extinction correction: *SHELXL*
Extinction coefficient: 0.015 (2)

Table 1
Selected geometric parameters (Å, °).

O1—C10	1.245 (5)	C9—C11	1.376 (6)
O2—C20	1.206 (5)	C9—C10	1.441 (6)
N3—C11	1.321 (5)	C18—C20	1.490 (6)
N3—C18	1.444 (5)	C18—C19	1.518 (6)
C7—C9	1.440 (6)		
C11—C9—C10—O1	4.1 (8)	N3—C18—C20—O2	−28.6 (7)

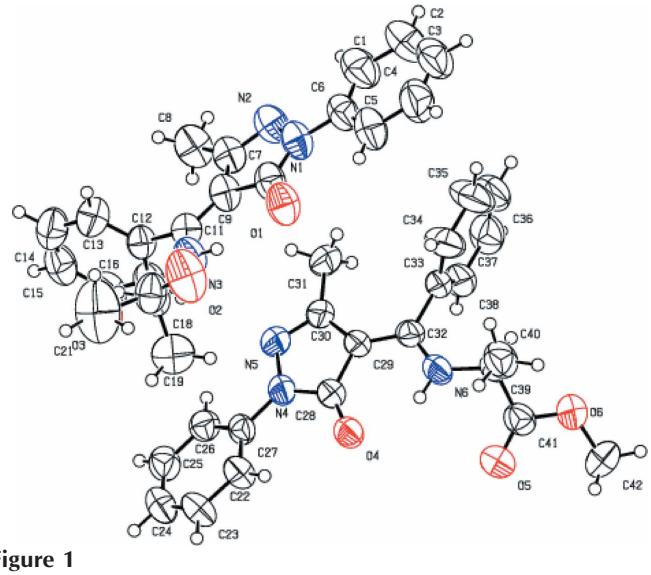


Figure 1
The asymmetric unit of the title compound. Displacement ellipsoids are drawn at the 50% probability level.

Table 2
Hydrogen-bond geometry (Å, °).

D—H···A	D—H	H···A	D···A	D—H···A
N3—H3···O1	0.86	2.00	2.713 (5)	139
N6—H6···O4	0.86	2.01	2.715 (4)	138

H atoms were positioned geometrically and treated as riding, with C—H = 0.93–0.96 Å, N—H = 0.86 Å, and U_{iso}(H) = 1.2U_{eq}(C,*N*) [1.5U_{eq}(C) for methyl groups]. In the absence of significant anomalous scattering effects, Friedel pairs were merged.

Data collection: *APEX2* (Bruker, 2003); cell refinement: *APEX2*; data reduction: *SAINT* (Bruker, 2001); program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *SHELXTL* (Bruker, 2001); software used to prepare material for publication: *SHELXTL*.

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