between non-H atoms are given in Table 2. The phenyl ring is planar to within experimental accuracy. The amide group is also planar, with deviations from the best least-squares plane of N(1) +0·018 (4), C(5) −0·014 (4), H(N1) −0·01 (4), H′(N1) −0·00 (4), O(2) +0·007 (3) Å. The amide moiety folds back and forms a 'seven-atom ring' with the open end closed by an intramolecular hydrogen bond [O(1)···O(2) = 2·683 (4), H(O1)–O(1) = 0·84 (4), H(O1)···O(2) = 1·88 (4) Å, O(1)–H(O1)···O(2) = 160 (4)°]. The C(5)–O(2) acceptor bond of 1·245 (3) Å and the C(2)–O(1) donor bond of 1·430 (4) Å are in good agreement, to within experimental accuracy, with the expected bond lengths caused by hydrogen-bond formation (Jeffrey, Ruble, McMullan, DeFrees, Binkley & Pople, 1980).

A stereopicture of the extended structure is shown in Fig. 2. The dihedral angle between phenyl rings of neighbouring molecules is 47 (1)°. The carbon chains in the unit cell lie approximately along **c**. This arrangement favours a weak intermolecular hydrogen bonding between molecules of neighbouring unit cells [N(1)···O(2) = 2·925 (5), H(N1)–N(1) = 1·02 (4), H(N1)···O(2) = 1·91 (4) Å, N(1)–H(N1)···O(2) = 174 (3)°].

The distance between the centroid of the aromatic ring and the electron-donor oxygen [O(2)], believed to be of significance in the biological activity of the molecule, is 6·10 (1) Å, a value in the range found for phenacemide (6·39 Å) and ethylphenacemide (5·85 Å), two potent anticonvulsant agents (Camerman & Camerman, 1977).

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The Structures of 3-Allyl-9-benzoyl-2-methyl-9-phenyl-4,9-dihydropyrazolo[5,1-*b*]quinazoline, $C_{27}H_{23}N_3O$, and 6,7-Dimethyl-1,3,3-triphenyl-1*H*-imidazo[1,2-*b*]pyrazol-2(3*H*)-one, $C_{25}H_{21}N_3O$

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Abstract. $C_{27}H_{23}N_3O$ (2*b*): $M_r = 405{\cdot}5$, orthorhombic, *Pbca*, $a = 13{\cdot}025$ (4), $b = 15{\cdot}991$ (3), $c = 20{\cdot}593$ (6) Å at 297 K, $V = 4289$ (2) Å^3, $Z = 8$, $D_x = 1{\cdot}26$ g cm^{-3}, λ(Mo $K\alpha$) = 0·7107 Å, $\mu = 0{\cdot}64$ cm^{-1}, $F(000) = 1712$; R_F for 1671 unique reflections $[I \geq 2\sigma(I)] = 0{\cdot}0728$. $C_{25}H_{21}N_3O$ (3*a*), $M_r = 379{\cdot}5$, monoclinic, $P2_1/c$, $a = 11{\cdot}234$ (4), $b = 7{\cdot}021$ (1), $c = 25{\cdot}330$ (6) Å, $\beta = 91{\cdot}83$ (3)° at 299 K, $V = 1997$ (1) Å^3, $Z = 4$, $D_x = 1{\cdot}26$ g cm^{-3}, λ(Mo $K\alpha$) = 0·7107 Å, $\mu = 0{\cdot}70$ cm^{-1}, $F(000) = 800$; R_F for 1383 unique reflections $[I \geq 2{\cdot}75\sigma(I)] = 0{\cdot}0770$. Bond distances and angles are all within the expected ranges. The 1,2-diazacyclopentadiene rings in both structures are nearly planar.

Introduction. The antineoplastic activity of the imidazo[1,2-*b*]pyrazole ring system has received considerable attention during the past few years (Shoemaker, Ayers, D'Anna & Cysyk, 1981; Cory & Fleischer, 1980; Allen & Thornthwaite, 1980). The known medicinal activity of fused pyrazoles has spurred considerable research into the synthesis of imidazo[1,2-*b*]pyrazoles (Elnagdi, Hafez & El-Fahham, 1980; Pilgram, 1980; Elguero, Jacquier & Mignonac-Mondon, 1973; Schulze & Willitzer, 1967) as well as the pyrazolo[5,1-*b*]quinazolines (Sircar, Capiris, Kesten & Herzig, 1981; Puetter, Wolfrum & Menzee, 1963) which appear in this paper. It has been suggested that the antitumor

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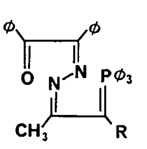
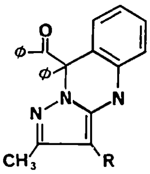
activity of 9,10-dihydroacridines, which are structurally related to (2), may be correlated to the dihedral angle formed by the folding of the central, six-membered ring; the flatter the ring, the greater the activity (Hempel, Hull, Bogucka-Ledochowska & Danter, 1979). Electrocyclic reactions of azines and α,β-unsaturated azines are well known (Schweizer & Evans, 1978; Wagner-Jauregg, 1976). We have also shown that cumulated azines are excellent synthons for a variety of fused pyrazoles.

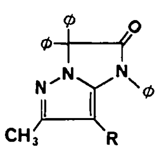
The phosphorane (1) on treatment with phenyl isocyanate (in a 1 to 1·25 ratio) in refluxing benzene solution for 4 h yields a mixture of (2) and (3) separable by column chromatography. Compound (2) was recrystallized from anhydrous ether and (3) from ethanol. We herein report the X-ray crystallographic characterization of (2*b*) and (3*a*). A full account of the preparation, spectroscopic properties and mechanistic considerations will be available elsewhere (Schweizer & Lee, 1984).



(1*a*) $R = CH_3$
(1*b*) $R = CH_2{-}CH{=}CH_2$



(2*a*) $R = CH_3$
(2*b*) $R = CH_2{-}CH{=}CH_2$



(3*a*) $R = CH_3$
(3*b*) $R = CH_2{-}CH{=}CH_2$

Experimental. Both structures: Nicolet *R*3 diffractometer at 296 K, $\theta/2\theta$ scan technique. The angular settings of 25 reflections ($20° \leq 2\theta \leq 30°$) determine unit-cell parameters. Atomic scattering factors from *International Tables for X-ray Crystallography* (1974). Throughout, computer programs were *P*3, *SHELXTL* (Sheldrick, 1981) and *XP* (Nicolet Corporation). Space groups unique from systematic absences, solution by direct-methods routine *SOLV* and difference Fourier syntheses. Final refinement (on *F*), all non-hydrogen atoms anisotropic, hydrogens fixed, idealized (dC—H = 0·96 Å) updated locations.

(2*b*) A finely formed, thin, colorless, brick-like crystal, 0·08 × 0·30 × 0·40 mm, affixed to a glass fiber. Of 2712 reflections ($3{\cdot}0 \leq 2\theta \leq 42{\cdot}0°$, $+h$, $+k$, $+l$), 2430 unique, and 1671 [$I \geq 2\sigma(I)$] used for structure solution and refinement. Three standard reflections ($\bar{1}15$, $\bar{3}02$, 220) every 141 reflections, no decay. Blocked-cascade refinement, 280 parameters, $R = 0{\cdot}0728$, $R_w = 0{\cdot}0680$, $S = 1{\cdot}304$, $g = 0{\cdot}0008$ [$w^{-1} = \sigma^2(F) + gF^2$], $(\Delta/\sigma)_{\max} = 0{\cdot}176$, final difference-map density $-0{\cdot}276$–$0{\cdot}305$ e Å^{-3}.

(3*a*) An irregular thick blade cleaved from a larger specimen affixed to a glass pin. Of 2762 reflections ($3{\cdot}0 < 2\theta \leq 43{\cdot}0°$, $+h$, $+k$, $\pm l$), 2312 unique, and 1383 [$I \geq 2{\cdot}75\sigma(I)$] used in structure solution and refinement. Three standard reflections (10,0,0, 080, 4,0,10) every 141 reflections, no decay. Refinement of 262 parameters, $R = 0{\cdot}0770$, $R_w = 0{\cdot}0821$, $S = 1{\cdot}053$, $g = 0{\cdot}001$, $(\Delta/\sigma)_{\max} = 0{\cdot}368$, final difference-map density $-0{\cdot}270$–$0{\cdot}273$ e Å^{-3}.

Discussion. Atomic coordinates are given in Tables 1 (2*b*) and 3 (3*a*).*

Table 1. *Atom coordinates* ($\times 10^4$) *and temperature factors* (Å$^2 \times 10^3$) *for* (2*b*)

	x	*y*	*z*	U_{eq}*
N(1)	4273 (3)	4088 (2)	2412 (2)	44 (1)
N(4)	6912 (3)	4585 (3)	2253 (2)	47 (1)
N(10)	5136 (3)	4399 (2)	2108 (2)	39 (1)
O	3647 (3)	5040 (2)	827 (2)	62 (1)
C(2)	4636 (4)	3848 (3)	2995 (2)	44 (2)
C(3)	5691 (4)	3988 (3)	3068 (2)	40 (2)
C(5)	7838 (4)	5530 (3)	1549 (2)	47 (2)
C(6)	7865 (4)	6053 (3)	1021 (2)	56 (2)
C(7)	7013 (4)	6160 (3)	638 (2)	59 (2)
C(8)	6140 (4)	5719 (3)	776 (2)	54 (2)
C(9)	5137 (3)	4637 (3)	1422 (2)	39 (2)
C(11)	5980 (3)	4335 (3)	2490 (2)	37 (2)
C(12)	6951 (3)	5102 (3)	1699 (2)	39 (2)
C(13)	6088 (3)	5174 (3)	1304 (2)	39 (2)
C(14)	3935 (4)	3446 (3)	3479 (2)	61 (2)
C(15)	6334 (4)	3830 (4)	3654 (2)	58 (2)
C(16)	6225 (5)	4445 (4)	4193 (3)	68 (3)
C(17)	5644 (5)	5096 (5)	4196 (3)	89 (3)
C(18)	4183 (3)	5178 (3)	1301 (2)	46 (2)
C(19)	3915 (4)	5894 (3)	1746 (2)	44 (2)
C(20)	2915 (4)	6191 (3)	1715 (3)	60 (2)
C(21)	2617 (4)	6851 (3)	2110 (3)	77 (2)
C(22)	3320 (6)	7217 (4)	2518 (3)	77 (3)
C(23)	4309 (5)	6941 (4)	2538 (3)	71 (3)
C(24)	4601 (4)	6274 (3)	2160 (2)	58 (2)
C(25)	5133 (4)	3843 (3)	982 (2)	41 (2)
C(26)	5901 (4)	3694 (4)	538 (2)	59 (2)
C(27)	5889 (5)	2987 (4)	154 (3)	71 (3)
C(28)	5106 (5)	2414 (4)	221 (2)	70 (3)
C(29)	4340 (4)	2552 (4)	662 (2)	66 (2)
C(30)	4356 (4)	3265 (3)	1044 (2)	57 (2)

* Equivalent isotropic *U* defined as one third of the trace of the orthogonalized U_{ij} tensor.

Table 2. *Bond distances* (Å) *for* (2*b*) *with e.s.d.'s*

O—C(18)	1·220 (6)	C(5)—C(12)	1·379 (7)
N(1)—C(2)	1·346 (6)	C(6)—C(7)	1·372 (7)
N(1)—N(10)	1·379 (5)	C(7)—C(8)	1·368 (7)
N(4)—C(11)	1·368 (6)	C(8)—C(13)	1·395 (6)
N(4)—C(12)	1·408 (5)	C(9)—C(13)	1·527 (6)
N(10)—C(9)	1·463 (5)	C(9)—C(18)	1·534 (6)
N(10)—C(11)	1·355 (6)	C(9)—C(25)	1·559 (6)
C(2)—C(3)	1·400 (7)	C(12)—C(13)	1·393 (6)
C(2)—C(14)	1·497 (7)	C(15)—C(16)	1·489 (8)
C(3)—C(11)	1·367 (6)	C(16)—C(17)	1·288 (10)
C(3)—C(15)	1·491 (7)	C(18)—C(19)	1·508 (7)
C(5)—C(6)	1·371 (7)	Ave. aromatic C—C	1·377 (8)

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* Lists of structure factors, anisotropic thermal parameters, H-atom coordinates and bond angles have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 39091 (26 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 3. *Atom coordinates* ($\times 10^4$) *and temperature factors* ($Å^2 \times 10^3$) *for* (3*a*)

	x	*y*	*z*	U_{eq}*
O	7423 (4)	9605 (7)	3326 (2)	53 (2)
N(1)	5771 (4)	7897 (7)	3569 (2)	35 (2)
N(4)	6653 (4)	5570 (8)	4032 (2)	38 (2)
N(5)	6541 (5)	4069 (8)	4379 (3)	44 (2)
C(2)	6975 (6)	8316 (10)	3571 (3)	37 (3)
C(3)	7647 (5)	6892 (9)	3948 (3)	32 (2)
C(6)	5361 (6)	3851 (10)	4386 (3)	40 (3)
C(7)	4697 (6)	5187 (10)	4079 (3)	36 (3)
C(7a)	5579 (6)	6264 (9)	3872 (3)	32 (2)
C(8)	4854 (7)	2269 (10)	4703 (3)	55 (3)
C(9)	3375 (6)	5467 (12)	4048 (3)	57 (3)
C(10)	4868 (5)	8786 (10)	3241 (3)	34 (2)
C(11)	4632 (7)	10697 (11)	3289 (3)	50 (3)
C(12)	3706 (7)	11477 (12)	2981 (3)	61 (3)
C(13)	3027 (7)	10356 (11)	2636 (3)	56 (3)
C(14)	3288 (6)	8459 (11)	2592 (3)	47 (3)
C(15)	4212 (6)	7678 (11)	2900 (3)	43 (3)
C(16)	8078 (5)	7793 (10)	4460 (3)	36 (3)
C(17)	7869 (6)	9683 (11)	4584 (3)	51 (3)
C(18)	8198 (7)	10438 (12)	5071 (3)	60 (3)
C(19)	8744 (7)	9289 (13)	5436 (4)	62 (4)
C(20)	8976 (6)	7399 (12)	5326 (3)	59 (3)
C(21)	8640 (5)	6645 (11)	4842 (3)	39 (3)
C(22)	8649 (6)	5928 (10)	3656 (3)	37 (3)
C(23)	8659 (6)	4040 (10)	3524 (3)	40 (3)
C(24)	9574 (7)	3293 (12)	3233 (3)	61 (3)
C(25)	10490 (7)	4439 (13)	3076 (3)	70 (4)
C(26)	10498 (7)	6311 (12)	3216 (4)	69 (4)
C(27)	9614 (6)	7076 (12)	3504 (3)	56 (3)

* Equivalent isotropic *U* defined as one third of the trace of the orthogonalized U_{ij} tensor.

Table 4. *Bond distances* (Å) *for* (3*a*) *with e.s.d.'s*

O–C(2)	1·216 (9)	C(2)–C(3)	1·561 (10)
N(1)–C(2)	1·384 (8)	C(3)–C(16)	1·509 (10)
N(1)–C(10)	1·433 (8)	C(3)–C(22)	1·525 (9)
N(1)–C(7a)	1·401 (8)	C(6)–C(7)	1·415 (10)
N(4)–C(3)	1·472 (8)	C(6)–C(8)	1·493 (10)
N(4)–N(5)	1·381 (8)	C(7)–C(7a)	1·364 (9)
N(4)–C(7a)	1·352 (8)	C(7)–C(9)	1·498 (9)
N(5)–C(6)	1·335 (9)	Ave. aromatic C–C	1·379 (11)

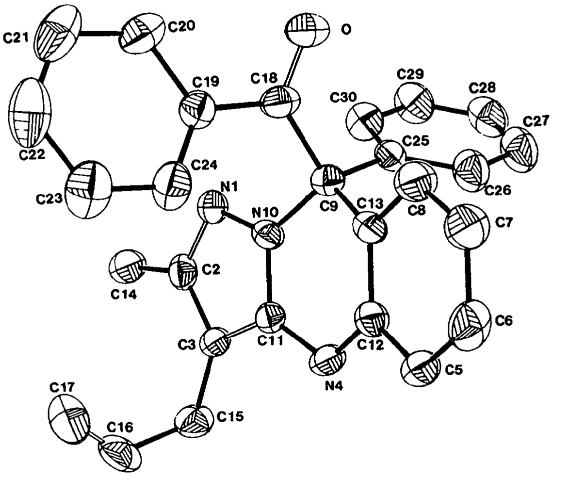

Fig. 1. *ORTEP* diagram (Johnson, 1965) and molecular labelling scheme for (2*b*). Ellipsoids are drawn at the 50% level and H atoms have been omitted.

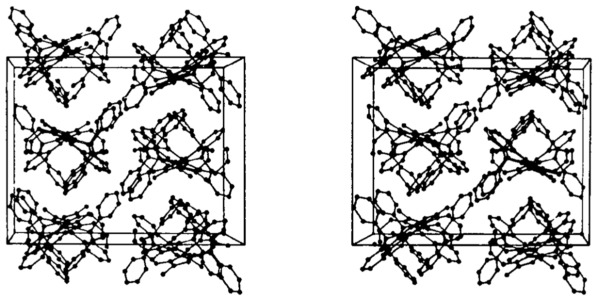

Fig. 2. Stereoview of the unit-cell packing of (2*b*) as viewed along the *a* axis.

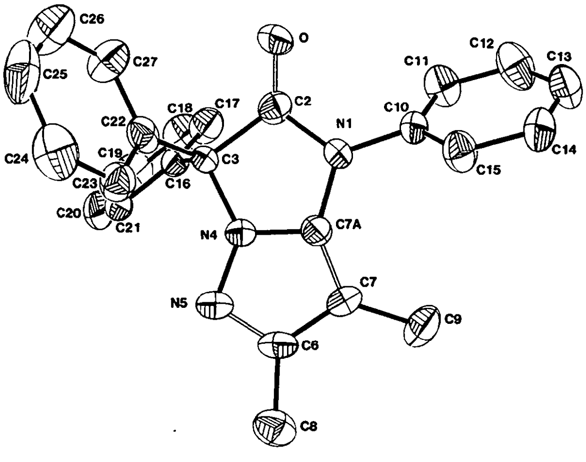

Fig. 3. *ORTEP* diagram (Johnson, 1965) and molecular labelling scheme for (3*a*). Ellipsoids are drawn at the 50% level and H atoms have been omitted.

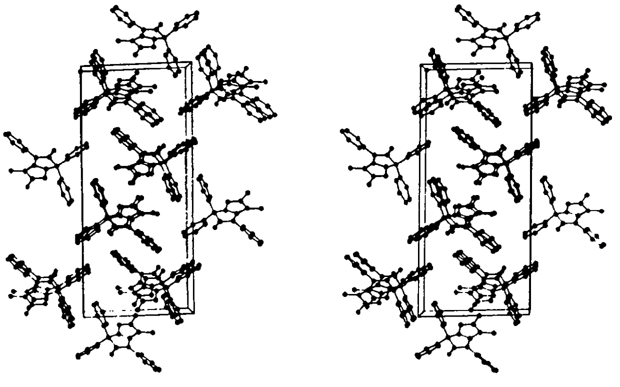

Fig. 4. Stereoview of the unit-cell packing of (3*a*) as viewed along the *b* axis.

Both (2*b*) and (3*a*) crystallize as well separated individual molecules with the structures shown in Figs. 1 and 3; packing diagrams are provided in Figs. 2 and 4. Bond distances for (2*b*) and (3*a*) are provided in Tables 2 and 4, respectively; these parameters are all within the expected ranges for the elements, bond orders and ring sizes found in both molecules. The 1,2-diazacyclopentadiene rings in both are nearly planar.

The most noteworthy feature of either molecule is the dihedral angle in (2*b*) in the central six-membered ring related by a line joining N(4) and C(9). The folding angle found is 157·0 (6)°. This can be compared to the analogous angle in 9,10-dihydroanthracene, 145° (Ferrier & Iball, 1954), *trans*-2-methyl-4,9-diphenyl-4,9-dihydropyrazolo[1,5-*b*]isoquinoline, 170·7 (4)° (Schweizer, Hsueh, Rheingold & Durney, 1983), 9-isopropyl-9,10-dihydroacridine, 154·2 (5)°* (Chu, Rosenstein & Napoleone, 1980), and 9-*tert*-butyl-9,10-dihydroacridine, 168·9 (5)°† (Chu & Rosenstein, 1979). Calculations have suggested that *trans*-9,10-disubstituted 9,10-dihydroanthracenes are non-planar (Rabideau, 1978; Dalling, Zilm, Grant, Heeschen, Horton & Pugmire, 1981); while unsubstituted 9,10-dihydroanthracene (Lipkowitz *et al.*, 1982) and 1,4-cyclohexadiene (Birch, Hinde & Radom, 1981) are planar. NMR studies on *N*-substituted 9,10-dihydroacridines have suggested that the molecule is in a flattened boat shape with a relatively low barrier for inversion (Aizenshtat, Klein, Weiler-Feilchenfeld & Bergmann, 1972; Taylor & Proctor, 1969, 1971).

In (3*a*), the dihedral angle relating the two five-membered rings is 8·5 (6)°.

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* Values calculated from reported coordinates.

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