question comes from examination of the intermolecular hydrogen-bonding network involved in the crystal packing which agrees with the residual electron density found on two carboxyl residues and interpreted as COOH H atoms.

Indeed, the crystal is stabilized by $-CO\cdots HOH\cdots OC-$ and $-COOH\cdots OOC-$ hydrogen bonds (Table 3) running respectively parallel and perpendicular to the macrocycles. Thus C(1) and C(8) bear carboxylic acid groups and, in order to keep the electronic balance, hydrazine must be in the monocationic form. Nevertheless, the importance of packing forces in crystallizing this particular complex as well as the existence of an $H_3\overset{+}{N}-\overset{+}{N}H_3$ complex at a slightly more acidic pH remain to be answered.

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CI Pigment Yellow 74, α-(2-Methoxy-4-nitrophenylhydrazono)-α-aceto-2′-methoxyacetanilide

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Abstract. 2-[(2-Methoxy-4-nitrophenyl)hydrazono]-*N*-(2-methoxyphenyl)-3-oxobutanamide, $C_{18}H_{18}N_4O_6$, $M_r = 386{\cdot}4$, triclinic, $P\bar{1}$, $a = 10{\cdot}729$ (2), $b = 11{\cdot}976$ (2), $c = 7{\cdot}628$ (1) Å, $\alpha = 103{\cdot}43$ (1), $\beta = 110{\cdot}28$ (1), $\gamma = 88{\cdot}06$ (1)°, $V = 892{\cdot}6$ (3) Å^3, $Z = 2$, $D_m = 1{\cdot}433$ (5), $D_x = 1{\cdot}4376$ (5) Mg m^{-3}, Cu $K\alpha$, $\lambda = 1{\cdot}54184$ Å, $\mu = 0{\cdot}83$ mm^{-1}, $F(000) = 404$, room temperature, $R = 7{\cdot}2\%$ for 3385 independent non-zero intensities. The molecule exists as the hydrazone tautomer and four intramolecular hydrogen bonds keep it approximately planar. There is one intermolecular hydrogen bond per molecule and this causes the compound to pack as dimers. The planar dimers are linked by van der Waals forces into columns.

Introduction. This article is one of a series (Whitaker, 1983*a*,*b*, 1984*a*,*b*, 1985*a*; Whitaker & Walker, 1985; Whitaker, 1985*b*, 1986) concerned with the structure of some acetoacetanilide azo-pigments, with a long-term aim of relating colour and crystal structure in pigments.

CI Pigment Yellow 74 (CI No. 11741) (I) is available commercially and is used in the manufacture of paint and printing inks. It was investigated to see what effect the two methoxy groups in the molecule have on the intramolecular hydrogen bonding. One of these groups is in the *ortho* position of the phenyl adjacent to the azo bond; this is unusual, as most commercial pigments of this type have a nitro group in this position. After the intensity data had been collected, the crystal structure

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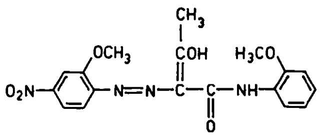
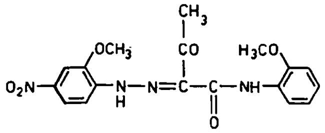
of another pigment, with two methoxy groups in the same positions but lacking the nitro group, was reported (Paulus, Rieper & Wagner, 1983).

Previous structure determinations in the series indicated that the term 'azo-pigments', as applied to this group of compounds, is a misnomer; the azo bond is, in fact, a hydrazone bond (II). This conclusion has been confirmed by other investigators (Paulus, Rieper & Wagner, 1983; Brown & Yadav, 1984; Paulus, 1984).



(I)



(II)

Experimental. The crystals were obtained by the slow cooling of a saturated hot (358 K) dimethylformamide solution of the commercial pigment AROT-043 Fastona Yellow 74.

The crystal measured 0·30 × 0·60 × 0·20 mm. Data were collected using an Enraf–Nonius CAD-4 automatic four-circle diffractometer of the National X-ray Diffractometer Service using filtered Cu radiation. Cell constants were obtained by least-squares analysis on 25 diffraction maxima with θ range 26·99 to 29·91°. The observed density was measured by flotation.

The observed intensities were corrected for Lorentz–polarization and absorption factors (North, Phillips & Mathews, 1968), maximum and minimum transmission factors 0·994 and 0·846, respectively, to give 3385 intensities up to a maximum 2θ of 140·0°, all independent and of non-zero intensity and all were used in the refinement. The ranges of Miller indices were $-13 \leq h \leq 13$, $-14 \leq k \leq 14$ and $0 \leq l \leq 9$. Three standard reflexions ($44\bar{4}$, $\bar{1}6\bar{1}$ and $4\bar{4}\bar{4}$) were measured periodically.

The structure was solved using the direct methods of *SHELXS*84 (after *SHELX*76 had failed). Refinement was based on F with weighting function $1/w = (\sigma^2 + 0{\cdot}03F^2)$; σ was obtained from the counting statistics.

After anisotropic refinement of the heavy atoms, a difference Fourier synthesis indicated the positions of the H atoms, confirming that the molecule was the hydrazone tautomer. The H atoms were included in the refinement with C—H and N—H distance 1·05 Å riding upon the appropriate heavy atom. Refinement continued until the shifts were less than one tenth the appropriate standard deviations (Mason, 1964). For the final refinement the methyl groups were freed from positional restraint. No attempt was made to refine the parameters of the other H atoms. Final $R = 0{\cdot}072$, $wR = 0{\cdot}090$. The comparatively high R value is attributed to the use of *all* data, which we consider to be methodologically more satisfactory than imposing an arbitrary cut-off (Hirshfeld & Rabinovich, 1973; Wilson, 1976, 1978). The final coordinates, together with equivalent isotropic temperature factors, are given in Table 1.* The maximum and minimum electron densities on the final difference Fourier synthesis were 0·36 and −0·61 e Å^{-3}, respectively.

Table 1. *Parameters and standard deviations after anisotropic refinement*

The equivalent isotropic temperature factor U_{eq} is defined by $\exp[-8\pi^2U(\sin^2\theta)/\lambda^2 \times 10^{-3}]$ (Cruickshank, 1956).

	x	y	z	U_{eq}(Å^2)
C(1)	0·9131 (2)	0·7576 (2)	0·5921 (2)	41 (1)
O(5)	0·9514 (1)	0·8591 (1)	0·5700 (2)	49 (1)
C(17)	1·0906 (2)	0·8818 (2)	0·6184 (4)	59 (1)
C(2)	0·9986 (2)	0·6793 (2)	0·6709 (3)	45 (1)
C(3)	0·9449 (2)	0·5793 (2)	0·6841 (3)	47 (1)
N(4)	1·0354 (2)	0·4985 (2)	0·7711 (3)	48 (1)
O(3)	1·1554 (2)	0·5197 (2)	0·8243 (3)	73 (1)
O(4)	0·9885 (2)	0·4108 (2)	0·7869 (3)	81 (2)
C(4)	0·8090 (2)	0·5543 (2)	0·6183 (3)	53 (1)
C(5)	0·7245 (2)	0·6330 (2)	0·5403 (3)	54 (1)
C(6)	0·7743 (2)	0·7350 (1)	0·5261 (3)	39 (1)
N(1)	0·6933 (2)	0·8167 (1)	0·4475 (2)	43 (1)
N(2)	0·5634 (1)	0·7937 (1)	0·3726 (2)	37 (1)
C(7)	0·4820 (2)	0·8655 (2)	0·2925 (3)	39 (1)
C(8)	0·3409 (2)	0·8198 (2)	0·2140 (3)	45 (1)
C(9)	0·3089 (2)	0·7081 (2)	0·2484 (4)	66 (1)
O(1)	0·2510 (1)	0·8721 (2)	0·1234 (3)	69 (1)
C(10)	0·5250 (2)	0·9788 (2)	0·2780 (3)	39 (1)
O(2)	0·6443 (1)	1·0121 (1)	0·3511 (2)	54 (1)
N(3)	0·4280 (2)	1·0401 (1)	0·1864 (2)	47 (1)
C(11)	0·4378 (2)	1·1499 (2)	0·1526 (3)	39 (1)
C(12)	0·3151 (2)	1·1923 (2)	0·0544 (3)	48 (1)
O(6)	0·2073 (1)	1·1212 (1)	0·0109 (3)	60 (1)
C(18)	0·0786 (2)	1·1589 (3)	−0·0841 (4)	67 (1)
C(13)	0·3118 (3)	1·2988 (2)	0·0131 (3)	58 (1)
C(14)	0·4284 (3)	1·3644 (2)	0·0695 (4)	64 (1)
C(15)	0·5476 (3)	1·3251 (2)	0·1667 (4)	66 (1)
C(16)	0·5533 (2)	1·2165 (2)	0·2072 (3)	55 (1)

Examination of the observed and calculated structure factors indicated that some intense reflexions (*e.g.* 110; $1\bar{2}1$; $\bar{1}12$ and $\bar{2}02$) were affected by extinction. No correction was applied for this and these structure factors were included in the refinement.

The following programs were used: *SHELXS*84 (Sheldrick, 1984) for the solution of the structure, *SHELX*76 (Sheldrick, 1976) for data processing, Fourier synthesis, least-squares refinements and atomic scattering factors, *BONDLA* for the calculation of atomic distances and angles, *PARST* (Nardelli, 1982)

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* Lists of structure factors, anisotropic thermal parameters, H-atom parameters, heavy-atom peak heights and intermolecular distances < 3·50 Å have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 44189 (22 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

for calculation of weighted least-squares planes of the molecule. Figs. 1 and 2 were drawn by *PLUTO* (Motherwell & Clegg, 1978).

Discussion. The bond lengths and interatomic angles are given in Table 2. Comparison of the observed bond lengths with accepted bond lengths (*International Tables for X-ray Crystallography,* 1968) confirms that the hydrazone tautomer is the better model, agreeing with previous determinations. Fig. 1 gives the molecular structure projected on to the plane containing the atoms C(1), N(3) and C(13).

The interatomic distances N(1)···O(2), N(1)···O(5), N(3)···O(1) and N(3)···O(6) suggest that there is a bifurcated hydrogen bond from N(1) to O(2) and O(5) and another from N(3) to O(1) and O(6) (Fig. 1). The first of these has been found in previously determined structures with similar values of the bond lengths, although most pigments have a nitro group rather than a methoxy group in the *ortho* position. Change of this group does not appear to affect the intramolecular hydrogen bonding. These hydrogen bonds inhibit rotation about neighbouring bonds and tend to hold parts of the molecule planar.

Although the molecule is almost planar, most atoms show significant deviations from this plane (Fig. 1). Both unsubstituted benzene groups are planar within 3·3σ but they are inclined at an angle of 5·68 (7)° with respect to each other. This angle compares with that of 37·65 (8)° for the other dimethoxy compound studied (Paulus, Rieper & Wagner, 1983, value recalculated).

There appear to be no abnormal bond lengths, although some differ by more than three times the standard deviation from those obtained in the other dimethoxy structure (Paulus, Rieper & Wagner, 1983). The most noticeable are the three nominal C—C single bonds; C(7)—C(8), C(8)—C(9), C(7)—C(10). In the present structure these are 1·492 (3), 1·494 (4) and 1·488 (3) Å, respectively (*i.e.* equal), while for the previously determined structure they are 1·462 (3), 1·501 (3) and 1·477 (3) Å, respectively, suggesting significant distortion in the bonding of the latter. This may be why this molecule is not planar while the present one is nearly so.

The molecules lie in columns parallel to the *c* axis; adjacent molecules within the columns are antiparallel. Unlike most other pigment structures that have been determined, the molecules in the columns are not directly above each other and the edges of the columns are puckered. This is unusual as the only other structures of this type in which the molecules do not sit in unpuckered columns are the other dimethoxy compound (Paulus, Rieper & Wagner, 1983) and CI Pigment Yellow 3 (Whitaker, 1983*a*).

The normal to the weighted molecular plane is 26·48 (2)° off-set with respect to the column axis; this plane makes an angle of 5·60 (1)° with the ($\bar{1}12$) plane.

Table 2. *Bond lengths* (Å) *and interatomic angles* (°)

C(17)—O(5)	1·428 (3)	C(7)—C(10)	1·488 (3)
O(5)—C(1)	1·362 (3)	C(10)—O(2)	1·243 (2)
C(1)—C(2)	1·379 (3)	C(10)—N(3)	1·339 (3)
C(2)—C(3)	1·386 (3)	N(3)—C(11)	1·412 (3)
C(3)—N(4)	1·452 (3)	C(11)—C(12)	1·414 (3)
N(4)—O(3)	1·225 (3)	C(12)—O(6)	1·357 (3)
N(4)—O(4)	1·228 (3)	O(6)—C(18)	1·432 (3)
C(3)—C(4)	1·385 (3)	C(12)—C(13)	1·380 (3)
C(4)—C(5)	1·376 (3)	C(13)—C(14)	1·380 (4)
C(5)—C(6)	1·392 (3)	C(14)—C(15)	1·368 (4)
C(6)—C(1)	1·411 (3)	C(15)—C(16)	1·399 (4)
C(6)—N(1)	1·387 (3)	C(16)—C(11)	1·378 (3)
N(1)—N(2)	1·321 (2)	N(1)—O(2)	2·597 (3)
N(2)—C(7)	1·304 (3)	N(1)—O(5)	2·623 (2)
C(7)—C(8)	1·492 (3)	N(3)—O(1)	2·657 (3)
C(8)—O(1)	1·224 (3)	N(3)—O(6)	2·568 (2)
C(8)—C(9)	1·494 (4)		

C(17)—O(5)—C(1)	117·1 (2)	C(10)—C(7)—N(2)	124·0 (2)
O(5)—C(1)—C(6)	114·7 (2)	C(7)—C(8)—C(9)	119·1 (2)
C(6)—C(1)—C(2)	120·2 (2)	C(9)—C(8)—O(1)	119·5 (2)
C(2)—C(1)—O(5)	125·1 (2)	O(1)—C(8)—C(7)	121·5 (2)
C(1)—C(2)—C(3)	118·5 (2)	C(7)—C(10)—N(3)	115·8 (2)
C(2)—C(3)—C(4)	122·5 (2)	N(3)—C(10)—O(2)	124·0 (2)
C(4)—C(3)—N(4)	119·3 (2)	O(2)—C(10)—C(7)	120·2 (2)
N(4)—C(3)—C(2)	118·2 (2)	C(10)—N(3)—C(11)	128·7 (2)
C(3)—N(4)—O(4)	118·6 (2)	N(3)—C(11)—C(12)	114·6 (2)
O(4)—N(4)—O(3)	122·5 (2)	C(12)—C(11)—C(16)	119·2 (2)
O(3)—N(4)—C(3)	118·9 (2)	C(16)—C(11)—N(3)	126·1 (2)
C(3)—C(4)—C(5)	118·6 (2)	C(11)—C(12)—O(6)	114·6 (2)
C(4)—C(5)—C(6)	120·8 (2)	O(6)—C(12)—C(13)	125·3 (2)
C(5)—C(6)—N(1)	122·9 (2)	C(13)—C(12)—C(11)	120·1 (2)
N(1)—C(6)—C(1)	117·6 (2)	C(12)—O(6)—C(18)	118·2 (2)
C(1)—C(6)—C(5)	119·4 (2)	C(12)—C(13)—C(14)	119·8 (2)
C(6)—N(1)—N(2)	118·6 (2)	C(13)—C(14)—C(15)	120·7 (2)
N(1)—N(2)—C(7)	121·6 (2)	C(14)—C(15)—C(16)	120·3 (2)
N(2)—C(7)—C(8)	112·0 (2)	C(15)—C(16)—C(11)	119·8 (2)
C(8)—C(7)—C(10)	124·0 (2)		

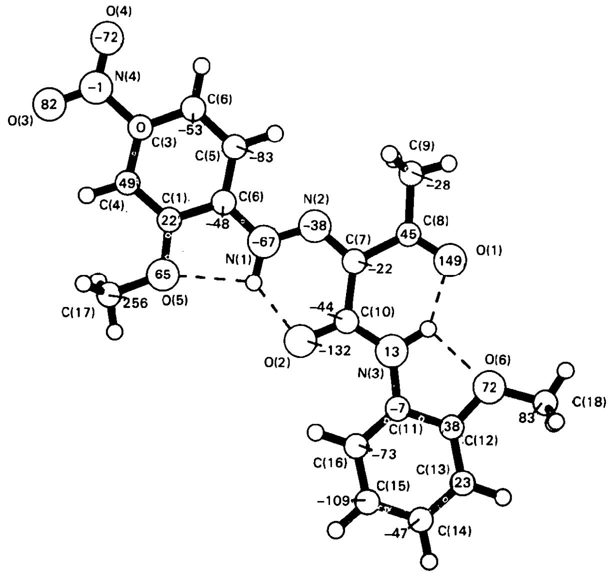

Fig. 1. Structure of the molecule projected on to the plane containing C(1), N(3) and C(13). The deviations of the atoms (in 10^{-3} Å) from the weighted least-squares plane of the molecules are also given. The errors in the atomic coordinates correspond to a mean of 0·002 Å. The intramolecular hydrogen bonds are indicated by broken lines.

The interplanar distance for $(\bar{1}12)$ planes is 3·318 Å and because there are substantial deviations from the molecular plane all intermolecular distances (excluding those involving hydrogen) below 3·50 Å were calculated (deposited). There are 22 of these of which 5 are between the columns (Fig. 2). One exceptionally short distance, C(17)···O(2) = 3·067 (3) Å, is much shorter than any other intermolecular distance [the second shortest is 3·382 (3) Å]. It can be compared with literature values for the van der Waals distance for methyl–oxygen of 3·40 Å (Pauling, 1960) and 3·28 Å [Bondi (1964); obtained by combining the methyl–methyl contact distance 3·57 Å with his van der Waals radius for oxygen, 1·50 Å]. The present distance is significantly shorter than either of these, suggesting the presence of an intermolecular CH_3···O hydrogen bond.

Sutor (1963) investigated short intermolecular and intramolecular C···O contacts, mainly in the region of 3·1 Å, which could be due to C—H···O hydrogen bonds, although, in many of the compounds considered, the positions of the H atoms had not been directly determined. Using the criterion that the H···O distance should be less than 2·6 Å [the sum of the van der Waals radii (Pauling, 1960)], a C···O distance as long as 3·28 Å was attributed to a hydrogen bond. It was found that in the case of methylidene (CH) groups, the C—H···O configuration was approximately linear and the H···O distance less than 2·6 Å, but for methylene (CH_2) and methyl (CH_3) groups, the angle C—H···O varied from 102 to 147° and the distance H···O could be larger than 2·6 Å. It was suggested that this may be caused by steric interference with the other H atoms of the methylene or methyl groups.

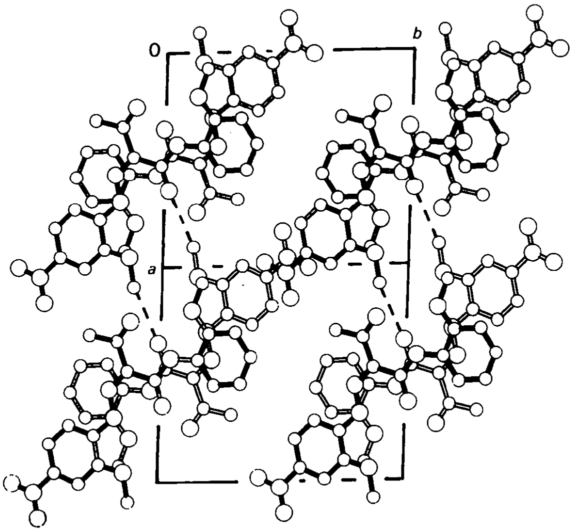

Fig. 2. The packing of CI Pigment Yellow 74 molecules in the unit cell, *c*-axis projection. The molecules at a higher level in the cell (*i.e.* those with larger *z* coordinates) are indicated by solid bonds. Two pairs of intermolecular hydrogen bonds showing two dimers are indicated by broken lines.

Table 3. *Intermolecular distances and angles about the methyl group* C(17)H_3

	x	*y*	*z*
C(17)	1·0905 (2)	0·8818 (2)	0·6184 (4)
H(171)	1·147 (3)	0·886 (2)	0·748 (4)
H(172)	1·092 (3)	0·949 (3)	0·584 (4)
H(173)	1·113 (3)	0·817 (3)	0·538 (4)
O(2)	1·3557 (1)	1·0514 (1)	0·4160 (2)

Distances (Å)		Angles (°)	
C(17)···O(2)	3·067 (3)	C(17)–H(171)···O(2)	86 (2)
C(17)–H(171)	0·96 (3)	C(17)–H(172)···O(2)	103 (2)
C(17)–H(172)	0·90 (3)	C(17)–H(173)···O(2)	78 (2)
C(17)–H(173)	0·95 (3)		
H(171)···O(2)	2·97 (3)		
H(172)···O(2)	2·74 (3)		
H(173)···O(2)	3·12 (3)		

Examination of the distances and angles about the methyl group (Table 3) strongly suggests an intermolecular hydrogen bond, C(17)—H(172)···O(2), approximately parallel to the *a* axis, with H···O 2·74 (3) Å.

This means that the molecular packing within the crystal is not that of individual molecules but of overlapping planar dimers. It seems that this dimer formation destroys the usual columnar nature of the acetoacetanilide pigment structure. This is similar to CI Pigment Yellow 3 (Whitaker, 1983*a*) where the molecules are held in sheets by intermolecular hydrogen bonds of length 3·079 (14) Å. For α-acetyl-2′-methoxy-α-(2-methoxyphenylhydrazono)anilide, Paulus, Rieper & Wagner (1983) state: 'There are no possibilities for intermolecular hydrogen bonds', but a recalculation identifies a short intermolecular distance similar to that in the present compound, C(17)···O(1) = 3·174 (4), C(17)—H(171) = 1·029 (31), H(171)···O(1) = 2·571 (29) Å and C(17)—H(171)···O(1) = 117 (2)°. This bond is still shorter than the van der Waals distance for methyl–oxygen and also supports the thesis that the common columnar packing is modified by intermolecular hydrogen bonding.

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CI Pigment Yellow 65, α-(4-Methoxy-2-nitrophenylhydrazono)-α-aceto-2′-methoxyacetanilide

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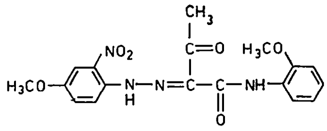
Abstract. 2-[(4-Methoxy-2-nitrophenyl)hydrazono]-*N*-(2-methoxyphenyl)-3-oxobutanamide, $C_{18}H_{18}N_4O_6$, M_r = 386·37, monoclinic, $P2_1/c$, a = 7·2949 (5), b = 8·9333 (5), c = 27·5600 (16) Å, β = 94·302 (5)°, V = 1791·0 (0·2) Å^3, Z = 4, D_x = 1·4330 (2), D_m = 1·431 (3) Mg m^{-3}, $F(000)$ = 808, Cu $K\alpha$, λ = 1·5418 Å, μ = 0·83 mm^{-1}, R = 9·5% for 3047 independent non-zero intensities. The molecule exists as the hydrazone tautomer and four intramolecular hydrogen bonds keep it approximately planar. With the usual criterion there are two intermolecular hydrogen bonds per molecule but one is very weak.

Introduction. This article is one of a series (Whitaker, 1983*a,b*, 1984*a,b*, 1985*a,b*, 1986; Whitaker & Walker, 1985, 1987) concerned with the long-term aim of relating colour and crystal structure in acetoacetanilide pigments.

Previous structure determinations have indicated that the term 'azo-pigments', as applied to this group of pigments, is a misnomer; the azo bond is in fact a hydrazone bond (I). This conclusion has been confirmed by other investigators (Paulus, Rieper & Wagner, 1983; Brown & Yadav, 1984; Paulus, 1984). It had also been found that in pigments of this type the pigment molecule contains intramolecular hydrogen bonding and this generally tends to keep the molecule approximately planar. The one exception to this generalization (Paulus, Rieper & Wagner, 1983) contained methoxy side groups. The present pigment is an isomer of a previously determined one, CI Pigment Yellow 74 (Whitaker & Walker, 1987). The pigment is commercially available and is used in oil media, especially for latex paints for exterior use.



(I)

Experimental. Single crystals were obtained by the slow cooling of a hot saturated toluene solution (358 K) of the commercial pigment, Fanchon Yellow RN YH5790. Optical examination indicated that the crystals were monoclinic.

A crystal of size 0·60 × 0·22 × 0·15 mm was used for intensity measurements. These were collected on an Enraf–Nonius CAD-4 automatic four-circle diffractometer (Cu $K\alpha$ radiation). Least-squares analysis of 25 diffraction maxima with θ range 16·04–29·75° gave the cell constants. Space group $P2_1/c$ with b parallel to the blade face and perpendicular to the length, a parallel to the length of the crystal and c approximately perpendicular to the blade face. The observed density was measured by flotation.

The intensities were corrected for absorption [(North, Phillips & Mathews, 1968), linear absorption coefficient 0·83 mm^{-1}, maximum and minimum transmission factors 0·997 and 0·854 respectively] and

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