990 (m), 915 (s), 805 (s), 775 (s), 747 (s), 705 cm^{-1} (s). 9-Methylpyrido[1,2-*a*]indole (**21f**) was separated as unstable yellow plates from pentane; mp 101.5–102.5 °C; NMR (CS_2) δ 2.43 (3 H, s, CH_3), 6.39 (1 H, dd, J = 7, 7 Hz, 7-H), 6.50 (1 H, s, 10-H), 6.58 (1 H, ddd, J = 1, 7, 7 Hz, 8-H), 7.07–7.42 (2 H, m, 2-, 3-H), 7.60–7.80 (2 H, m, 1-, 4-H), 8.12 (1 H, dd, J = 1, 7 Hz, 6-H); IR (KBr) 3050 (m), 2930 (m), 1625 (m), 1605 (m), 1515 (s), 1465 (s), 1350 (s), 1335 (m), 1305 (m), 1248 (m), 1225 (m), 1153 (s), 1118 (m), 1010 (m), 928 (m), 872 (m), 768 (s), 740 cm^{-1} (s); UV (EtOH) λ_{max} 218 (ϵ 9330), 230 (10 700), 256 (52 500), 260 (sh), 275 (sh), 288 (sh), 304 (2790), 314 (3470), 329 (1950), 355 (sh), 372 (2090), 388 (2630), 410 (2400), 434 nm (1050); high-resolution MS, *m/e* calcd for $C_{13}H_{11}N$, 181.0889, found, 181.0888. 8-Methylpyrido-[1,2-*a*]indole (**21g**) was separated as unstable yellow plates from pentane; mp 151–151.5 °C dec; NMR (CS_2) δ 2.29 (3 H, s, CH_3), 6.20 (1 H, dd, J = 2, 7 Hz, 7-H), 6.33 (1 H, s, 10-H), 6.93–7.28 (3 H, m, 2-, 3-, 9-H), 7.68–7.94 (2 H, m, 1-, 4-H), 8.07 (1 H, d, J = 7 Hz, 6-H); IR (KBr) 3040 (m), 2905 (m), 1635 (s), 1605 (s), 1520 (s), 1475 (s), 1460 (s), 1375 (m), 1345 (s), 1325 (s), 1240 (s), 1227 (s), 1172 (s), 1030 (m), 1007 (m), 985 (m), 920 (s), 860 (s), 770 (s), 740 (s), 725 (s), 700 cm^{-1} (s); UV (EtOH) λ_{max} 218 (ϵ 11 100), 230 (13 900), 257 (58 600), 262 (57 600), 277 (sh), 290 (sh), 304 (4390), 315 (5550), 330 (6340), 357 (sh), 378 (2440), 396 (2870), 417 (2360), 445 nm (sh). Anal. Calcd for $C_{13}H_{11}N$: C, 86.16; H, 6.12; N, 7.73. Found: C, 86.19; H, 6.13; N, 7.66. 7-Methyl-pyrido[1,2-*a*]indole (**21h**) was separated as yellow plates from pentane; mp 137.5–138 °C; NMR (CS_2) δ 2.26 (3 H, s, CH_3), 6.41 (1 H, s, 10-H), 6.58 (1 H, dd, J = 2, 9 Hz, 8-H), 6.96–7.28 (3 H, m, 2-, 3-, 9-H), 7.46–7.68 (2 H, m, 1-, 4-H), 7.78 (1 H, d, J = 1 Hz, 6-H); IR (KBr) 3050 (m), 2930 (m), 1605 (s), 1540 (s), 1520 (s), 1485 (m), 1460 (s), 1455 (s), 1420 (s), 1340 (s), 1325 (m), 1310 (s), 1265 (m), 1245 (s), 1237 (s), 1175 (s), 1135 (m), 1100 (m), 1035 (m), 1010 (m), 980 (m), 930 (s), 905 (s), 840 (m), 805 (s), 770 (s), 740 (s), 710 cm^{-1} (s); UV (EtOH) λ_{max} 217 (sh), 231 (ϵ 8710), 259 (49 000), 279 (sh), 292 (sh), 306 (2510), 320 (3020), 332 (1780), 362 (sh), 379 (2000), 399 (2450), 422 (2090), 449 nm (sh). Anal. Calcd for $C_{13}H_{11}N$: C, 86.16; H, 6.12; N, 7.73. Found: C, 86.06; H, 6.01; N, 7.47. 6-Methylpyrido[1,2-*a*]indole (**21i**) was separated as yellow plates from pentane; mp 57–59 °C dec; NMR (CS_2) δ 2.95 (3 H, s, CH_3), 6.13 (1 H, dd, J = 1, 6 Hz, 7-H), 6.53 (1 H, s, 10-H), 6.68 (1 H, dd, J = 7, 9 Hz, 8-H), 6.94–7.30 (3 H, m, 2-, 3-, 9-H), 7.55 (1 H, dd, J = 2, 6 Hz, 1- or 4-H), 8.04 (1 H, dd, J = 1, 7 Hz, 4- or 1-H); IR (KBr) 3030 (m), 1630 (s), 1595 (s), 1530 (s), 1473 (m), 1460 (m), 1435 (s), 1405 (s), 1378 (m), 1340 (m), 1305 (s), 1287 (s), 1250 (m), 1215 (s), 1150 (s), 1050 (m), 1032 (m), 1017 (m), 985 (s), 947 (s), 920 (m), 830 (s), 770 (s), 740 (s), 715 (s), 695 cm^{-1} (s); UV (EtOH) λ_{max} 217 (ϵ 12 900), 252 (68 500), 253 (sh), 260 (47 600), 278 (sh), 291 (sh), 305 (3610), 315 (4870), 330 (8170), 348 (sh), 368 (3270), 386 (3660), 408 (3250), 433 nm (1400). Anal. Calcd for $C_{13}H_{11}N$: C, 86.16; H, 6.12; N, 7.73. Found: C, 86.08; H, 6.23; N, 7.61.

Pyrolysis of 2-(1-Propenyl)pyridine *N*-Oxide (26).[30] The principal compound obtained from column chromatography (hexane–ether/alumina) of fraction I was indolizine (**28**) obtained as colorless plates from pentane; mp 71.5–72.5 °C (lit.[31] mp 73–74 °C), 162 mg (48%); NMR[32] δ 6.12–6.77 (4 H, m, 1-,2-,6-, 7-H), 7.14 (1 H, d, J = 1 Hz, 3-H), 7.25 (1 H, dd, J = 2, 8 Hz, 8-H), 7.73 (1 H, dd, J = 1, 8 Hz, 5-H); IR (KBr) 1625 (m), 1520 (m), 1450 (m), 1363 (s), 1318 (s), 1310 (s), 1243 (s), 1220 (m), 1150 (m), 1075 (s), 1035 (m), 765 (s), 735 (s), 715 cm^{-1} (s); UV[33] (H_2O) λ_{max} 232 (ϵ 36 300), 274 (4590), 280 (5400), 292 (6290), 336 nm (3030).

Pyrolysis of 2-(*o*-Tolyl)pyridine *N*-Oxide (27).[34] 4-Azafluorene (**29**) was obtained as plates from pentane, mp 93–94 °C (lit.[25] mp 95–97 °C, 69%) and 2-(*o*-tolyl)pyridine (11%) were isolated from fraction I. Toluene (11%) and pyridine (trace) were detected in fraction II.

Registry No. 1a, 694-59-7; **1b**, 931-19-1; **1c**, 1003-73-2; **1d**, 1003-67-4; **2a**, 110-86-1; **2b**, 109-06-8; **2c**, 108-99-6; **2d**, 108-89-4; **3**, 38746-50-8; **4**, 586-98-1; **5**, 100-71-0; **6**, 100-69-6; **7**, 1132-37-2; **8**, 4916-40-9; **9**, 1437-15-6; **12**, 82198-70-7; **14**, 101-82-6; **15**, 92-52-4; **16**, 2116-62-3; **17**, 103-29-7; **20a**, 20531-86-6; **20b** picrate, 82198-71-8; **20c** picrate, 82198-72-9; **20d**, 20531-88-8; **20e**, 80772-89-0; **20f**, 82198-73-0; **20g**, 80772-88-9; **20h**, 80772-87-8; **20i**, 80772-86-7; **21a**, 245-43-2; **21c**, 80772-77-6; **21d**, 80772-76-5; **21e**, 80772-84-5; **21f**, 80772-83-4; **21g**, 80772-82-3; **21h**, 80772-81-2; **21i**, 80772-80-1; **22b**, 36995-45-6; **22c**, 29263-64-7; **22d**, 29335-87-3; **22e**, 14159-54-7; **22f**, 56664-26-7; **22g**, 5191-54-8; **22h**, 63065-67-8; **22h** picrate, 82198-74-1; **22i**, 10131-46-1; **23**, 260-36-6; **25**, 15260-65-8; **26**, 21715-31-1; **27**, 33421-20-4; **28**, 274-40-8; **29**, 244-99-5; 4-methylpyrido[1,2-*a*]indole, 80772-78-7; 2-ethylpyridine *N*-oxide, 4833-24-3; 2-(*o*-tolyl)pyridine, 10273-89-9.

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Stereochemistry of Aroylphosphonate Phenylhydrazones and Their Conversion to 1*H*-Indazole-3-phosphonates

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Received February 5, 1982

Reaction of phenylhydrazine and (2,4-dinitrophenyl)hydrazine with dialkyl aroylphosphonates gives exclusively the *Z* isomers of the arylhydrazones **5**. Heating **5** in acetic acid produces an equilibrium mixture of **5** and the *E* isomers **6**. Oxidation of phenylhydrazones **a–g** (either **5** or **6**) with lead tetraacetate leads to azoacetates **7a–g**, which can be cyclized with BF_3–etherate to 1-phenyl-1*H*-indazole-3-phosphonates **8a–g**.

Aroylphosphonates **1**, which are valuable synthetic intermediates,[1] can react with nucleophiles **2** in either of two ways. Nucleophiles with an α heteroatom such as hydroxylamine[1a] and substituted hydrazines[2] condense with the carbonyl group to provide the corresponding oximes or hydrazones. With simple nucleophiles like water,[3a] alcohols,[3b] thiols,[3c] or amines,[1b,3d] acyl derivatives **4** are

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(2) (a) Berlin, K. D.; Taylor, H. A. *J. Am. Chem. Soc.* **1964**, *86*, 3862. (b) Kudzin, Z. H.; Kotynski, A. *Synthesis* **1980**, 1028. (c) Regitz, M.; Anschutz, W.; Liedhegener, A. *Chem. Ber.* **1968**, *101*, 3734.
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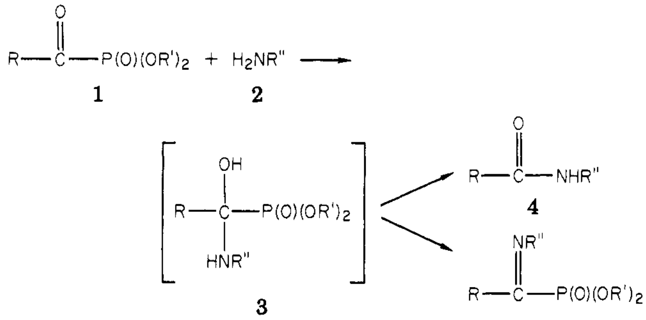
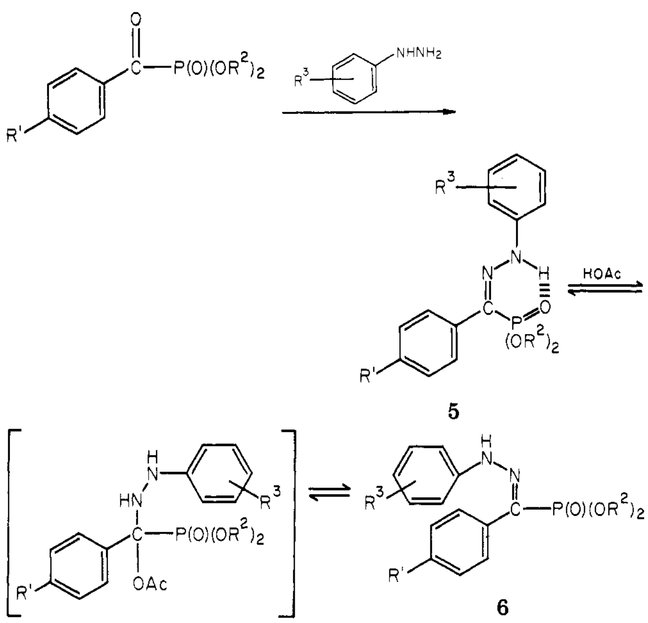
0022-3263/82/1947-3503$01.25/0

Scheme I



Scheme II[a]



[a] a, R^1 = H, R^2 = CH_3, R^3 = H; b, R^1 = OCH_3, R^2 = CH_3, R^3 = H; c, R^1 = OCH_3, R^2 = C_2H_5, R^3 = H; d, R^1 = Cl, R^2 = CH_3, R^3 = H; e, R^1 = Cl, R^2 = C_2H_5, R^3 = H; f, R^1 = CH_3, R^2 = CH_3, R^3 = H; g, R^1 = CH_3, R^2 = C_2H_5, R^3 = H; h, R^1 = H, R^2 = C_2H_5, R^3 = 2,4-$(NO_2)_2$; i, R^1 = OCH_3, R^2 = C_2H_5, R^3 = 2,4-$(NO_2)_2$.

formed by cleavage of the carbon–phosphorus bond. Both types of products can be formed through an intermediate such as **3** (**Scheme I**).

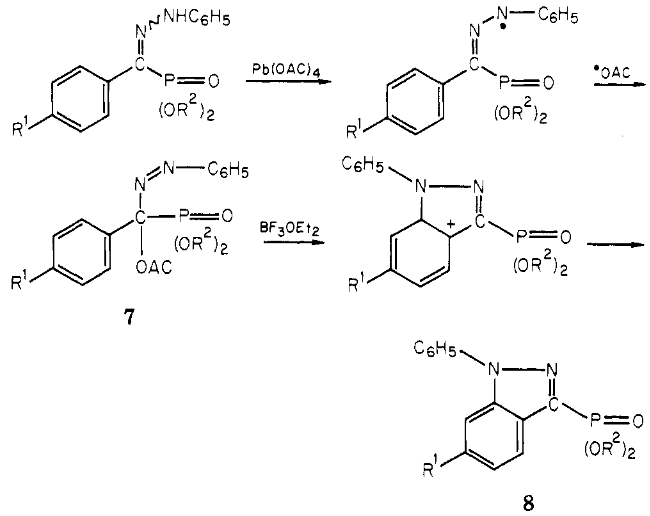
We have now investigated the reaction of aroylphosphonates with phenylhydrazine to examine the stereochemistry of the products and their conversion into 1*H*-indazolephosphonates. Although the reaction of dimethyl benzoylphosphonate with hydrazine hydrate is complex,[5] reaction of dialkyl aroylphosphonates with phenylhydrazines provides the phenylhydrazones in good yields (Scheme II). The gross structure of **5** was deduced from the absence of IR carbonyl absorption at 1650 cm^{-1} and the appearance of absorption at 1600 cm^{-1}. Elemental analyses and ^{1}H NMR spectra are also compatible with this structure (see Table I of the supplementary material).

Stereochemistry of Hydrazones

Arylhydrazones can exist in both *Z* (**5**) and *E* (**6**) configurations. The *Z* isomers should be stabilized by hydrogen bonding, whereas the *E* isomers should be less sterically crowded. Hydrazones **a**, **d**, **f**, **h**, and **i** were boiled

(5) Swamy, R. V., unpublished results.

Scheme III



in acetic acid, and the solutions were then chromatographed on a silica gel column, yielding two products that were shown to be isomeric by elemental analyses and spectral data (see Table II of the supplementary material). From the following evidence we conclude that the starting hydrazones are *Z* isomers (**5**) and that the second products formed on heating in acetic acid are the *E* isomers (**6**). The R_f values determined by TLC on silica gel plates are higher for **5** than for **6**. The greater mobility (and hence polarity) of **6** is substantiated by reverse-phase HPLC with methanol–water as the mobile phase, in which **6** appears before **5**. The IR spectra of **5** showed an O–H stretching band at ~3180 cm^{-1} that did not shift on dilution. In contrast, a concentrated solution of **6** showed IR bands at ~3320 and ~3200 cm^{-1}, the latter band disappearing on dilution. Furthermore, **5** (including **5h** and **5i**) show UV absorption at longer wavelengths than does **6**. Such differences in UV spectra have been seen in phenylglyoxalic acid hydrazones[6] and may be attributable to hydrogen bonding in the *Z* isomers. It appears that the phenyl hydrazones reported by Berlin and Taylor[2a] have the *Z* configuration. Our ^{1}H NMR spectra of **5** did not show any NH absorption, whereas such absorption was seen at δ 8.2 in the *E* isomers **6**.

Equilibrium between the two isomers could be established by heating either isomer in acetic acid. The ratios observed were **5a**/**6a** = 25:75, **5d**/**6d** = 28:72, **5f**/**6f** = 28:72, and **5h**/**6h** = 33:67. Exclusive formation of the **Z** isomers in the preparation is probably indicative of kinetic control. Since isomerization could not be effected by boiling in toluene, we conclude that it is acid catalyzed, proceeding by the pathway suggested in Scheme II.[7]

Oxidation of Hydrazones

The oxidation of phenylhydrazones with lead tetraacetate and the mechanism involved have been reported.[8] This oxidation followed by cyclization of the product constitutes a route to synthesis of heterocycles. We found that this oxidation could be carried out more conveniently in benzene than in acetic acid. Hydrazones **a**–**g** (either **5**

(6) Vogel, Ch.; Matter, M. *Helv. Chim. Acta* **1959**, *42*, 527. Ciganek, E. *J. Org. Chem.* **1970**, *35*, 862 (this reference also records longer wavelength absorption for glyoxalic ester hydrazones).

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or 6) were readily oxidized to azoacetates 7 (**Scheme III**), but the (2,4-dinitrophenyl)hydrazones **h–i** did not undergo oxidation.[8]

Azoacetate **7a** was characterized by correct elemental analysis and by spectral data. It did not have a strong IR band at 1600 cm^{-1} (C=N) but showed a band at 1740 cm^{-1} (ester C=O). In the ^{1}H NMR spectrum the methyl protons of the acetate group appeared at δ 2.1, and those of the phosphate ester appeared as a double doublet, indicating a nearby chiral center.

Boiling azoacetates **7a–g** with BF_3–etherate effected cyclization to 1*H*-indazoles **8a–g** in yields of 30–37%. The 1*H*-indazole structure is supported by the literature[8] as well as by elemental and spectral data (see Table III of the supplementary material and the experimental section). The ^{1}H NMR spectra of the aromatic protons are compatible with the 1*H*-indazole structure,[9,10a] and the UV absorption is similar to the 1*H*-indazole.[10b] The pathway shown in Scheme III is proposed for the formation of 8 from **5–6**. It is interesting that neither $Pb(OAc)_4$ nor BF_3–etherate cleaves the C–P bond, which is broken readily by MnO_2.[11]

Experimental Section

NMR spectra were recorded on a Perkin-Elmer R-32 instrument operating at 90 MHz. IR spectra were taken on a Perkin-Elmer Model 577 instrument in KBr pellets or neat (for liquids). Hydrogen-bonding studies were done in CCl_4 solutions. UV spectra were obtained with a Pye-Unicam 500 spectrophotometer on solutions in CH_3OH. HPLC analyses were performed on a Water Associates ALC/6PC/344 instrument with a C_{18} μ-Bondapack column. Melting points are uncorrected.

Dialkyl Aroylphosphonate Phenylhydrazones 5. Phenylhydrazine (0.1 mol) was added slowly (dropwise so that the reaction temperature did not exceed 30 °C) to a solution of dialkyl aroylphosphonate (0.1 mol) in 100 mL of ethanol. The mixture was stirred for 30 min at room temperature and allowed to stand overnight for the products to crystallize. The hydrazones were recrystallized from EtOH. **5a–g**: IR 3180–3200, 1600–1605, 1260 cm^{-1}; UV$_{max}$ (approximate log ϵ) 350–361 (4.4), 280–298 (3.9), 236–242 nm (4.3). **5h,i**: IR 3100, 1250–1260 cm^{-1}; UV$_{max}$ (approx log ϵ) 381–389 (4.5), 225–228 nm (4.2). Yield (%), mp (°C), and R_f (silica plates, 10% acetone in benzene) for 5 are as follows: **5a**, 80, 85, 0.68; **5b**, 70, 105, 0.66; **5c**, 67, 77–78, 0.71; **5d**, 75, 135, 0.68; **5e**, 71, 72–73, 0.75; **5f**, 75, 105–106, 0.67; **5g**, 73, 96, 0.77; **5h**, 75, 170, 0.75; **5i**, 80, 175, 0.72. Elemental analyses, IR, UV, and NMR data are given in Table I, of the supplementary material.

(*E*)-Phenylhydrazones 6. **5a, d, f, h**, or **i** was boiled with a 10-fold excess of acetic acid for 12 h, the solution was cooled and 100 mL of CH_2Cl_2 was added. This solution was washed with water and then with $NaHCO_3$. The solution was dried, the solvent evaporated, and the residue chromatographed on a silica gel column (benzene to elute **5** and 10% acetone in benzene to get **6**). The *E* isomers were recrystallized from EtOH. **6a,d,f**: IR 3320–3340, 3200–3220, 1230–1235 cm^{-1}; UV$_{max}$ (approxmiate log ϵ) 308–322 (4.2), 279–290 nm (4.0). **6h–i**: IR 3280, 3100, 1250 cm^{-1}; UV$_{max}$ (approx log ϵ) 343–347 (4.3), 253 nm (4.0); mp (°C) and R_f (same condition as in **5**) for **6** are as follows: **6a**, 110, 0.25; **6d**, 94, 0.29; **6f**, 108–109, 0.24; **6h**, 110, 0.37; **6i**, 120, 0.33. Elemental analyses, IR, UV, and NMR data are given in Table II of the supplementary material.

Azoacetates 7. A solution of **5** or **6** (0.1 mol) in 25 mL of dry benzene was added over 15 min to a stirred solution of 0.15 mol of $Pb(OAc)_4$ in 200 mL of dry benzene. The slightly exothermic reaction was maintained at 10 °C with an ice bath during the addition and then warmed to 20–25 °C and stirred for 30 min. The mixture developed a yellow color, and $Pb(OAc)_2$ precipitated. The mixture was stirred with 200 mL of water, and the PbO_2 was removed by filtration. The benzene layer was separated and washed successively with water and dilute $NaHCO_3$ until free of acetic acid. After the mixture was dried over $MgSO_4$, the benzene was removed and the product isolated.

1-Phenyl-1*H*-indazole-3-phosphonates 8. Crude **7** (0.1 mol) was dissolved in 100 mL of benzene, and BF_3–etherate (0.1 mol) was added. After heating under reflux for 1 h, the cooled mixture was poured into water. The benzene layer was washed with dilute $NaHCO_3$ to remove acetic acid and then dried over $MgSO_4$. The solvent was distilled, and the residue was chromatographed on silica gel to give **8**. **8a–g**: IR 1590–1600, 1245–1260, 1020–1025 cm^{-1}; UV$_{max}$ (approximate log ϵ) 289–302 (3.9), 242–248 (4.1), 205–212 nm (4.4) (this last band was missing in **8a**). **8d,f,g** had an additional band at 278 nm (3.8). Melting points for **8a** and **8b** are 72 and 75 °C, respectively. **8c–g** are liquids. The *m/e* for **8a** is 302, and that for **8g** is 344. Elemental analyses, IR, UV, and NMR data are given in Table III of the supplementary material.

Direct Preparation of 8. The dialkyl aroylphosphonate (0.1 mol) was added slowly to a solution of $Pb(OAc)_4$ (0.15 mol) in 200 mL of dry benzene. After 2 h with continuous shaking, the BF_3–etherate complex (0.1 mol) was added. After heating under reflux for 1 h, the cooled mixture was washed with water and dilute $NaHCO_3$ and dried over $MgSO_4$. The product was isolated by chromatography on silica gel (10% acetone in benzene as eluent).

Acknowledgment. We thank Drs. P. K. Ramachandran and S. K. Das for critical comments and we thank J. D. Sapre for preparation of the manuscript.

Registry No. 1 (R = Ph; R^1 = CH_3), 18106-71-3; 1 (R = *p*-$MeOC_6H_4$; R^1 = CH_3), 10570-48-6; 1 (R = *p*-$MeOC_6H_4$; R^1 = C_2H_5), 16703-95-0; 1 (R = *p*-ClC_6H_4; R^1 = CH_3), 33493-32-2; 1 (R = *p*-ClC_6H_4; R^1 = C_2H_5), 10570-46-4; 1 (R = *p*-MeC_6H_4; R^1 = CH_3), 33493-30-0; 1 (R =*p*-MeC_6H_4; R^1 = C_2H_5), 2942-54-3; 1 (R = Ph; R^1 = C_2H_5), 3277-27-8; **5a**, 72973-99-0; **5b**, 82228-61-3; **5c**, 82228-62-4; **5d**, 82228-63-5; **5e**, 82228-64-6; **5f**, 82228-65-7; **5g**, 82228-66-8; **5h**, 82228-67-9; **5i**, 82228-68-0; **6a**, 72974-00-6; **6d**, 82228-69-1; **6f**, 82228-70-4; **6h**, 82228-71-5; **6i**, 82228-72-6; **7a**, 82228-73-7; **7b**, 82228-74-8; **7c**, 82228-75-9; **7d**, 82228-76-0; **7e**, 82228-77-1; **7f**, 82228-78-2; **7g**, 82228-79-3; **8a**, 82228-80-6; **8b**, 82228-81-7; **8c**, 82228-82-8; **8d**, 82228-83-9; **8e**, 82228-84-0; **8f**, 82228-85-1; **8g**, 82228-86-2; phenylhydrazine, 100-63-0; 2,4-dinitrophenylhydrazine, 119-26-6.

Supplementary Material Available: Complete UV, IR, elemental analyses, and NMR data for compounds **5**, **6**, and **8** in Tables I–III (4 pages). Ordering information is given on any current masthead page.

(9) For example the ^{1}H NMR of **8a** has the following absorptions: δ 8.17 (d, J = 8 Hz, 1 H, H$_4$) 7.55 (m, 8 H), 3.95 (d, J = 10 Hz, 6 H, OCH_3).

(10) (a) Dennler, E. B.; Portal, C. R.; Frasca, A. R. *Spectrochim. Acta, Part A* **1967**, *23A*, 2243. (b) Rousseau, V.; Lindwall, H. G. *J. Am. Chem. Soc.* **1950**, *72*, 3047.

(11) Kaushik, M. P., unpublished results.