Identification of Products from Photolysis of 9. Methyl 3-methyl-3-(isopropoxy-1-*d*)butanoate-2-*d* (11) gave the following: ν 1733 cm^{-1} (ester C=O); δ 1.07 [m, 6 H, $(CH_3)_2CD$-], 1.25 [s, 6 H, $(CH_3)_2C<$], 2.39 (m, 1 H, -CHD-), and 3.61 (s, 3 H, OCH_3).

Anal. Calcd for $C_9H_{16}D_2O_3$: C, 61.33; H and D, 11.44. Found: C, 61.54; H and D, 11.34.

Methyl 3-methyl-2-butenoate-2-*d* (12) gave the following: ν 1718 (conjugated ester C=O) and 1643 cm^{-1} (C=C); δ 1.88 (s, 3 H, *trans*-CH_3C=CCO-), 2.15 (s, 3 H, *cis*-CH_3C=CCO-), and 3.62 (s, 3 H, OCH_3).

Anal. Calcd for $C_6H_9D_2O_2$: C, 62.53; H and D, 9.70. Found: C, 62.32; H and D, 10.08.

Registry No.—2, 5455-94-7; 3, 25859-48-7; 4, 924-50-5; 5, 25859-51-2; 6, 25859-52-3; 9, 25859-53-4; 11, 25907-97-5; 12, 25859-50-1.

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Benzene Ring Substituted Indeno[1,2-*c*]pyrazol-4(1*H*)-ones

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The condensation of 4-substituted 2-acetyl-1,3-indandiones (**1a–d**) with hydrazine yielded the corresponding indandione α-hydrazones (**2a–c**), indeno[1,2-*c*]pyrazol-4(1*H*)-ones (**3** and **6**) or indeno[1,2-*c*]pyrazol-4(1*H*)-one hydrazones (**7a–c**), depending upon the nature of the substituents and the reaction conditions. Evidence is presented that the product of the reaction of 2-acetyl-4-nitro-1,3-indandione (**1a**) with hydrazine is 3-methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one (**6**), whereas the product from 2-acetyl-4-amino-1,3-indandione (**1b**) and hydrazine is 5-amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (**3**) and that the ring closure of the α-hydrazones of 4-amino- and 4-acetamido-2-acetyl-1,3-indandione (**2a** and **2b**) yields 5-amino- and 5-acetamino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (**3** and **4**). 2-Acetyl-5-nitro-1,3-indandione (**12**) was treated with hydrazine to give 3-methyl-6- (or 7-) nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one hydrazone (**13**).

A number of indeno[1,2-*c*]pyrazol-4(1*H*)-ones with substituents in the pyrazole ring has been reported in several papers from this laboratory.[1,2] The interesting physiological properties of these compounds prompted us to investigate the syntheses and characteristics of benzene ring substituted 3-methylindeno[1,2-*c*]pyrazol-4 (1*H*)-ones **3**, **4**, **6**, **7**, and **13**, Scheme I).

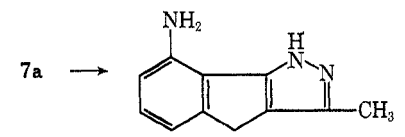
The preparation of 4- and 5-substituted 2-acetyl-1,3-indandiones (**1a–d** and **12**) necessary for this investigation was described in a previous paper.[3] The condensation of 4-amino- and 4-acetamido-2-acetyl-1,3-indandione (**1b** and **1c**) with 1 or 2 mol of hydrazine in refluxing ethanol for 0.5–1 hr yielded the α-hydrazones of the corresponding 2-acetylindandiones (**2a** and **2b**). When these hydrazones were refluxed in ethanol (in the case of **2a** catalytic amounts of hydrochloric acid were necessary) cyclization to the corresponding indeno-[1,2-*c*]pyrazol-4(1*H*)-ones **3** and **4** occurred. Whereas when **2a** and **2b** were refluxed in acetic anhydride the α-acetylhydrazone of 4-acetamido-2-acetyl-1,3-indandione (**5**) was formed.

The condensation of 4-amino-2-acetyl-1,3-indandione (**1b**) with hydrazine at 180–200° under the conditions of the Wolff–Kishner reaction gave directly 5-amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (**3**), whereas 4-nitro-2-acetyl-1,3-indandione (**1a**) yielded directly 8-nitro-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (**6**) by reacting with 1 mol of hydrazine in refluxing ethanol for 48 hr. When an excess of hydrazine was used, the nitroindandione **1a** gave the indenopyrazolone hydrazone **7a**. Under these last conditions 4-acetamido- and 4-hydroxy-2-acetyl-1,3-indandione (**1c** and **1d**) also gave the corresponding indenopyrazolone hydrazones **7b** and **7c**. All attempts to form the hydrazone of 5-amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one from **1b** failed.

The hydrazono group in the indandione hydrazones **2a–c** is on the side chain carbonyl, as demonstrated by a positive Tollens test[4] and by the formation of a red solution with aqueous sodium hydroxide.[4]

In the condensation of compounds **1** with hydrazine to form **3**, **6**, and **7** and in the cyclization of compounds **2** to form **3** and **4**, only one of the two possible isomers, the 5- or 8-substituted 3-methylindeno[1,2-*c*]pyrazol-4-(1*H*)-one, was formed. Theoretical considerations would lead one to predict that these reactions would give the 8-substituted rather than the 5-substituted isomer, when R is an electron-withdrawing group and the 5 rather than the 8 isomer, when R is an electron-donating group.

Evidence for the structure of the nitro derivative **7a** was obtained by treating it with hydrazine and palladium on charcoal to form the amino-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole (**8**). The melting point and infrared spectrum of **8** were found to be different from those of an authentic sample of 5-amino-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole (**10**, see below) and therefore the 8-nitro structure was assigned to com-



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(2) R. A. Braun and W. A. Mosher, *J. Org. Chem.*, **24**, 648 (1959).

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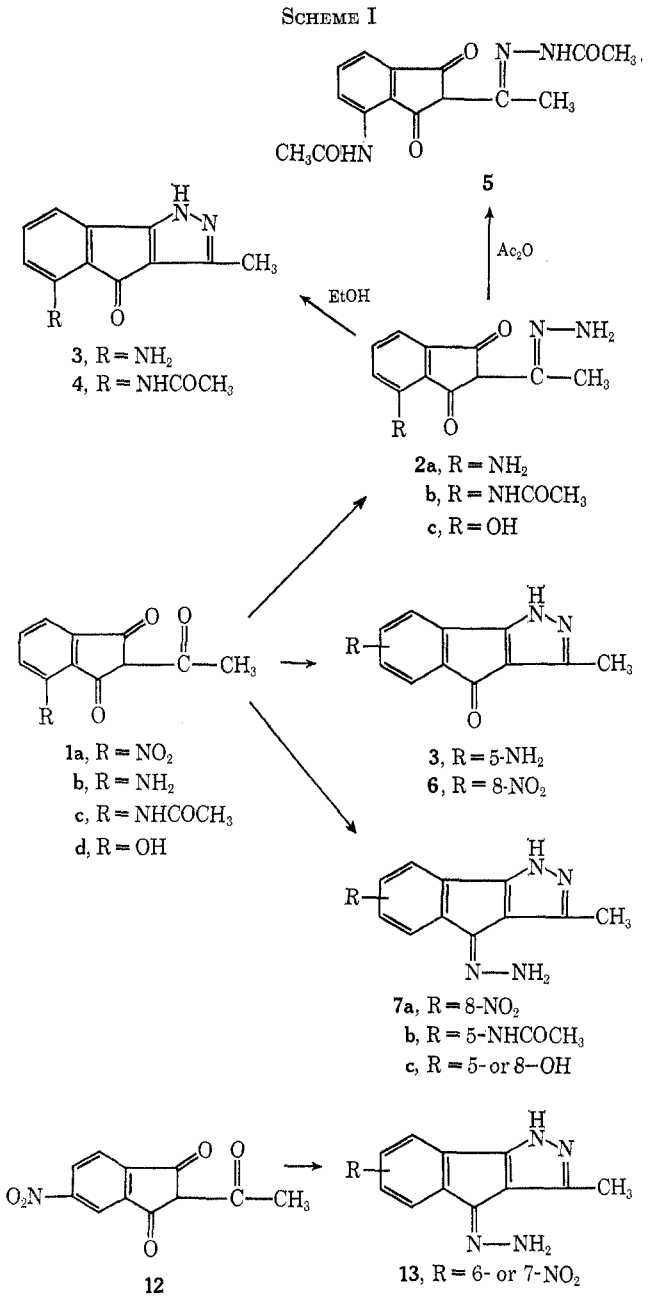
SCHEME I



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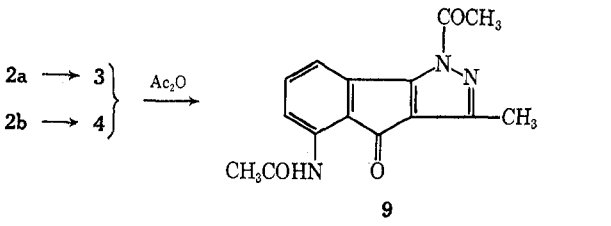
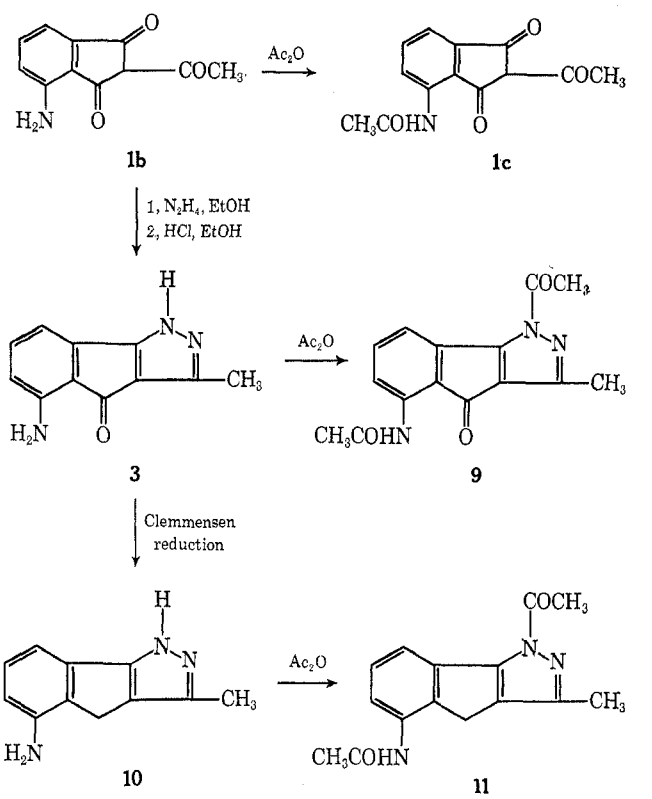


SCHEME III



pound **7a**. These results also support the structure assigned to compound **6**. Orientation of the acetamido group in **7b** was established by treating **7b** with hydrazine and potassium hydroxide under the conditions of the Wolff–Kishner reaction. The product obtained was identical with an authentic sample of 5-amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (**3**, see below).

Proof that the 5-substituted isomer was formed in the cyclization of compounds **2a** and **2b** was obtained by determining the positions of the amino and acetamido groups in compounds **3** and **4** respectively. First it was established that cyclization of **2a** and **2b** to form **3** and **4**, followed by acetylation (Scheme II), gave the same indenopyrazolone **9**, thus demonstrating that the amino group in **3** is in the same position as the acetamido group in **4**. Then compounds **1c**, **9**, and **11** were prepared as shown in Scheme III and their infrared spectra compared. These spectra show that the hydrogen atoms of the acetamido groups of **1c** and **9** exhibit similar intramolecular hydrogen–oxygen bonding (N—H stretching frequencies 3330 cm^{-1} for **1c** and 3335 cm^{-1} for **9**), whereas compound **11** exhibits an absorption frequency corresponding to a free N—H stretching (3435 cm^{-1}). These results demonstrate that the acetamido group in compound **9** must be in position 5 and therefore support the structures, 5-amino- and 5-acetamido-3-methyl[1,2-*c*]pyrazol-4-(1*H*)-one, assigned to compounds **3** and **4**, respectively. This structure proof of **3** is obviously also a proof for the structure **10** assigned to the amino-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole.

The condensation of 2-acetyl-5-nitro-1,3-indandione (**12**) with hydrazine in ethanol yielded directly the hydrazone of 3-methyl-6- (or 7-) nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one (**13**). Attempts to determine the position of the nitro group were unsuccessful.

The infrared spectra of the benzene ring substituted 3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-ones show a broad band at about 3200 cm^{-1} (N—H stretching of the pyrazole), a band at 1720 cm^{-1} (ketone carbonyl) when the benzene ring is substituted with a nitro group, and a band at 1690 cm^{-1} (ketone carbonyl) when the benzene ring is substituted with an amino or acetamido group. The band at 3200 cm^{-1} disappears upon acetylation of the pyrazole ring.

Experimental Section[5]

2-Acetyl-4-amino-1,3-indandione α-Hydrazone (2a).—Into a flask equipped with a mechanical stirrer and condenser were placed 50 ml of anhydrous ethanol and 0.7 g (0.02 mol) of 95% hydrazine. The mixture was heated to reflux and 2.03 g (0.01 mol) of 2-acetyl-4-amino-1,3-indandione[3] was added. The amine dissolved rapidly to give a red solution. After 7 min a yellow precipitate was formed. The mixture was refluxed for 1 hr, then was cooled and the precipitate was collected by filtration, washed with ethanol and dried to give 1.5 g (69%) of **2a** as small yellow needles, mp 220–222° (dimethylformamide–water).

Anal. Calcd for $C_{11}H_{11}N_3O_2$: C, 60.82; H, 5.12; N, 19.35. Found: C, 61.05; H, 5.46; N, 20.09.

Prolonged refluxing and/or a large excess of anhydrous hydrazine led to the same product.

4-Acetamido-2-acetyl-1,3-indandione α-Hydrazone (2b).—Hydrazine hydrate 100% (1.2 g, 0.025 mol) was added to a stirred, refluxing solution of 4-acetamido-2-acetyl-1,3-indandione[3] (5 g, 0.02 mol) in ethanol (300 ml). The yellow solution turned red and shortly thereafter a mass of bright yellow needles separated. One 0.5 hr after the addition of the hydrazine, the mixture was cooled and the precipitate was collected by filtration, washed with ethanol, and dried to yield 4.5 g (79%) of **2b**, mp 264–265.5 (dimethylformamide–water). When the melting point is taken slowly, the product melts above 300°, probably because it cyclizes to the corresponding indenopyrazolone.

Anal. Calcd for $C_{13}H_{13}N_3O_3$: C, 60.23; H, 5.02; N, 16.22. Found: C, 59.85; H, 5.19; N, 16.15.

2-Acetyl-4-hydroxy-1,3-indandione α-Hydrazone (2c).—Hydrazine hydrate 85% (1 ml, 0.022 mol) was added to a solution of 2-acetyl-4-hydroxy-1,3-indandione[3] (1.2 g, 0.006 mol) in ethanol (50 ml). After 5 min the precipitate was collected by filtration and washed with ethanol to give 0.7 g (55%) of **2c** as yellow needles, mp 260–262° dec (dimethylformamide–water).

Anal. Calcd for $C_{11}H_{10}N_2O_3$: C, 60.55; H, 4.59; N, 12.84. Found: C, 60.76; H, 4.67; N, 13.09.

The hydrazone **2c** was also obtained when a solution of 4-acetoxy-2-acetyl-1,3-indandione[3] (0.1 g) in anhydrous ethanol (10 ml) was refluxed for 10 min with 95+% hydrazine (2 drops).

4-Acetamido-2-acetyl-1,3-indandione α-(Acetylhydrazone) (5). A.—A small amount of **2a** was dissolved in acetic anhydride and the solution refluxed for several minutes whereupon a heavy precipitate of thin white needles formed. The precipitate was collected by filtration, washed with ether, and dried to give **5**, as creamy white needles, mp 263.5–264.5° (ethanol), in almost quantitative yield.

Anal. Calcd for $C_{15}H_{15}N_3O_4$: C, 59.80; H, 4.98; N, 13.95. Found: C, 59.79; H, 5.04; N, 13.92.

B.—The previous reaction was repeated using the acetamido **2b** in place of the amino compound **2a**. The product was identical with that obtained (part A above) by acetylation of **2a**, as shown by comparison of the infrared spectra and by mixture melting point determinations.

3-Methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one (6).—To a suspension of 2-acetyl-4-nitro-1,3-indandione[3] (5 g, 0.02 mol) in 250 ml of ethanol was added 10 g (0.02 mol) of a hydrazine solution, prepared by dissolving 13 g of 85% hydrazine hydrate in 87 g of ethanol. After stirring under reflux for 48 hr the reaction mixture was filtered hot to remove 2 g of suspended greenish-yellow solid and the filtrate was refrigerated for 18 hr to yield 2.2 g (50%) of golden yellow crystals. Sublimation at 220° (1.5 mm), followed by recrystallization from ethanol gave **6** as light yellow crystals of orange fluorescence, mp 251.5–252°.

Anal. Calcd for $C_{11}H_7N_3O_3$: C, 57.64; H, 3.06; N, 18.34. Found: C, 57.97; H, 3.07; N, 18.27.

5-Amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (3). A. From indandione 1b (Wolff–Kishner Reduction Conditions).—In an open flask 2.0 g (0.01 mol) of amine **1b**[3] and 2.0 g (0.04 mol) of 100% hydrazine hydrate were added to 30 ml of diethylene glycol; the solution was heated to 150° over a 1-hr period. Then 1 g of pulverized potassium hydroxide was added and the temperature quickly raised to 180°, held there for 1 hr and then raised to 200° for 15 min. To the reaction mixture 200 ml of water was added at room temperature and a small amount of a brown resinous substance filtered off and discarded. The filtrate was extracted with seven 100-ml portions of ether. The combined extracts were washed with 50 ml of water and dried over $MgSO_4$. Removal of the solvent left 1.1 g of yellow solid, which chromatographed on an alumina-packed column with tetrahydrofuran as the eluent gave 1.0 g (51%) of **3** as crystals of bright yellow fluorescence, mp 280–283°.

Anal. Calcd for $C_{11}H_9N_3O$: C, 66.33; H, 4.52; N, 21.10. Found: C, 66.24; H, 4.85; N, 20.81.

B. From Hydrazone 2a.—A suspension of compound **2a** (5 g, 0.025 mol) in anhydrous ethanol (300 ml) was heated to reflux and a few drops of concentrated hydrochloric acid were added. Within 2 min a clear solution was obtained. Refluxing was continued for an additional hour, and then the solvent was evaporated *in vacuo* leaving 5 g of crude orange material. Sublimation at 200° (1 mm) gave 4.2 g (85%) of **3** as pale yellow crystals, mp 280–283°, identical (melting point and ir) with **3** obtained by procedure A.

5-Acetamido-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (4).—A mixture of **2b** (4.5 g, 0.017 mol) and 200 ml of anhydrous ethanol was refluxed with stirring for 48 hr during which time most of the solid gradually dissolved. The reaction mixture was filtered while hot to remove 1.0 g of insoluble nonfluorescent material. Concentration of the filtrate gave 2.5 g (60%) of **4** as pale yellow crystals of yellow fluorescence, which upon sublimation at 200° (1 mm) and recrystallization from ethanol melted above 300°.

Anal. Calcd for $C_{13}H_{11}N_3O_2$: C, 64.73; H, 4.56; N, 17.42. Found: C, 64.78; H, 4.64; N, 17.39.

Compound **4** when refluxed in a small portion of ethanol containing a few drops of concentrated hydrochloric acid gave the amine **3** in quantitative yield.

5-Acetamido-1-acetyl-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one (9). A. From the Aminoindenopyrazolone 3.—To 1.0 g (0.0041 mol) of **3** in a test tube was added 2 ml (0.01 mol) of acetic anhydride. The mixture was heated slowly until the solid went into solution and then was refluxed for 5 min, and upon cooling to room temperature 1.0 g (77%) of bright yellow needles of diacetylated product was obtained, which upon sublimation at 170° (1 mm) and recrystallization from acetic anhydride gave **9** as yellow needles of bright yellow fluorescence and mp 206–208°.

Anal. Calcd for $C_{15}H_{13}N_3O_3$: C, 63.60; H, 4.59; N, 14.84. Found: C, 63.55; H, 4.70; N, 14.66.

B. From the Acetamidoindenopyrazolone 4.—A mixture of 1.0 g of **4** and 2 ml of acetic anhydride was refluxed for several minutes then cooled to room temperature. The precipitate was collected by filtration, washed, and purified to give yellow needles. A mixture melting point with a sample of **9** prepared by acetylation of **3** (part A above) showed no depression.

3-Methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one Hydrazone (7a).—A mixture of 2-acetyl-4-nitro-1,3-indandione[3] (3.0 g, 0.013 mol) and 95% hydrazine (2.5 g, 0.078 mol) in absolute ethanol (200 ml) was stirred and refluxed for 48 hr. Complete solution was never attained, but the color of the solid material in the flask changed from yellow to orange. The hot reaction mixture was filtered to give 2.6 g (82.5%) of **7a** as orange crystals, slightly soluble in most solvents. Soxhlet extraction with toluene or xylene for 24 hr gave orange-yellow needles, mp 270–273°.

Anal. Calcd for $C_{11}H_9N_5O_2$: C, 54.32; H, 3.70; N, 28.81. Found: C, 54.03; H, 3.91; N, 29.45.

To a refluxing mixture of hydrazone **7a** (0.5 g), dioxane (10 ml), and acetone (5 ml) were added 2 drops of concentrated hydrochloric acid and the refluxing continued for 0.5 hr. The solid was collected by filtration and crystallized from xylene to give a quantitative yield of 3-methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one *azine with acetone* as pale yellow, nonfluorescent needles, mp 297–299°.

Anal. Calcd for $C_{14}H_{13}N_5O_2$: C, 59.36; H, 4.59; N, 24.73. Found: C, 59.48; H, 5.00; N, 24.57.

Hydrazone **7a** and 2-nonanone, when refluxed as described above, gave 3-methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one *azine with 2-nonanone* as yellow, nonfluorescent crystals, mp 133–134° (ethanol).

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(5) Melting points were determined with a Fisher–Johns melting point apparatus and are uncorrected. The melting points of the hydrazones were taken by preheating the apparatus to a few degrees below the melting point of the samples. Then the samples were placed on the hot stage and the temperature was raised rapidly to the melting point. Infrared spectra were taken on a Perkin-Elmer Infracord spectrophotometer Model 137 using sodium chloride plates.

Samples used in H-bonding studies were dissolved in chloroform and the infrared spectra were taken on a Perkin-Elmer spectrophotometer Model 421. Elemental analyses were performed by Dr. A. Bernhardt Microanalytisch Laboratorium in Max Planck Institut für Kohlenforschung, Mülhein (Ruhr), West Germany.

Anal. Calcd for $C_{20}H_{25}N_5O_2$: C, 65.39; H, 6.81. Found: C, 64.83; H, 6.87.

5-Acetamido-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one Hydrazone (7b).—A mixture of 4-acetamido-2-acetyl-1,3-indandione[3] (5 g, 0.020 mol), anhyrous ethanol (400 ml), and 95+% hydrazine (6 ml, 0.19 mol) was stirred and refluxed for 48 hr. An orange solution was rapidly formed, and then a yellow solid precipitated which gradually redissolved. The reaction mixture was filtered hot to remove a small amount of grayish powder. The filtrate was cooled in a refrigerator for 12 hr and the precipitate was collected by filtration, washed with water, and dried to give 4.5 g (88%) of **7b** as pale yellow needles, mp 264.5–265° (ethanol).

Anal. Calcd for $C_{18}H_{18}N_6O$: C, 61.18; H, 5.10. Found: C, 61.21; H, 5.14.

Hydrazone **7b**, when treated with hydrazine and potassium hydroxide following the procedure described for compound **3** from **1b**, gave 5-amino-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one, identical with an authentic sample prepared as described above.

5- (or 8-) Hydroxy-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one Hydrazone (7c).—By the method used for the preparation of **7b**, a mixture of 2-acetyl-4-hydroxy-1,3-indandione[3] (3.0 g, 0.015 mol) in anhydrous ethanol (400 ml) was reacted with 95+% hydrazine (4 ml, 0.12 mol). A 15% yield (0.5 g) of **7c** was obtained as pale yellow crystals, mp 298–300° (ethanol) with darkening and sublimation.

Hydrazone **7c**, when refluxed in acetone for 5–10 min, gave 5- (or 8-) hydroxy-3-methylindeno[1,2-*c*]pyrazol-4(1*H*)-one *azine with acetone* as pale yellow needles, mp 299–300°.

Anal. Calcd for $C_{14}H_{14}N_4O$: C, 66.14; H, 5.51; N, 22.05. Found: C, 66.30; H, 5.57; N, 21.96.

3-Methyl-6- (or -7-) nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one Hydrazone (13).—To a stirred and refluxing mixture of 2-acetyl-5-nitro-1,3-indandione[3] (1.0 g, 0.004 mol) and absolute ethanol (100 ml), 95+% hydrazine (1.0 ml, 0.03 mol) was added. The indandione dissolved rapidly to give an orange-red solution from which a precipitate of yellow crystals began to form after approximately 2 hr. An additional 0.5 ml of 95% hydrazine was added and refluxing was continued for 22 hr. The precipitate was collected by filtration, washed with ethanol, and dried to give 0.84 g (89%) of **13** as yellow platelets, decomposing at about 300° (dimethylformamide–water).

Anal. Calcd for $C_{11}H_9N_5O_2$: N, 28.81. Found: N, 28.74.

5-Amino-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole (10).—A mixture of 10 g of zinc amalgam (prepared from mossy zinc, mercuric chloride, and concentrated hydrochloric acid[6]), 7.5 ml of water, 17.5 ml of concentrated hydrochloric acid, and 1 g (0.005 mol) of **3** was heated on a steam bath for 10 hr. The colorless solution still hot was then decanted from any unchanged zinc amalgam. The residue in the flask was extracted by boiling with 10 ml of water and the clear solution decanted. The combined decanted solutions were cooled and 1.2 g of colorless needles was collected by filtration. These crystals were dissolved in 100 ml of water, and the solution was made basic with 1 *N* NaOH and extracted with five 100-ml portions of ether. The combined ether extracts were dried over KOH pellets, filtered, and treated with excess dry hydrogen chloride. The precipitate was removed by filtration and dissolved in a small amount of water and the solution treated with enough 1 *N* KOH to precipitate the free amine. Filtration and washing of the precipitated amine with a small portion of cold water gave 0.52 g (56%) of **10**, as colorless needles, mp 224–226° (water).

Anal. Calcd for $C_{11}H_{11}N_3$: C, 71.35; H, 5.95; N, 22.70. Found: C, 71.40; H, 5.92; N, 22.65.

5-Acetamido-1-acetyl-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole (11).—Amine **10** (0.2 g) was added to acetic anhydride (1 ml) and the mixture heated to reflux for several minutes. The precipitate formed upon cooling was collected by filtration, washed with ether, and dried to give a quantitative yield of **11**. Sublimation at 220° (1 mm), followed by crystallization from acetic anhydride, gave **11** as needles of mp 260–262°.

Anal. Calcd for $C_{15}H_{15}N_3O_2$: C, 66.91; H, 5.58. Found: C, 66.82; H, 5.69.

8-Amino-1,4-dihydro-3-methylindeno[1,2-*c*]pyrazole (8).—A solution of 10 ml of 95+% hydrazine in 50 ml of ethanol was added dropwise over a 2.5-hr period to a stirred and refluxing mixture of **7a** (2.6 g, 1.1 mol) and 10% Pd–C (0.7 g) in ethanol (200 ml). The mixture was refluxed for additional 72 hr and then was filtered still hot, and the filtrate evaporated to dryness to give 1.2 g (60%) of crude **8**. Recrystallization from ethanol–water (Darco) gave **8** as pale yellow needles, mp 205.5–208°. The mixture melting point of this compound and the amine **10** (mp 224–226°) showed a significant depression (mmp 171–182°) and the ir spectra showed differences in the "fingerprint" region.

Anal. Calcd for $C_{11}H_{11}N_3$: C, 71.35; H, 5.95; N, 22.70. Found: C, 71.35; H, 6.21; N, 22.92.

Registry No.—**2a**, 25906-41-6; **2b**, 25906-42-7; **2c**, 25906-43-8; **3**, 25906-44-9; **4**, 25906-45-0; **5**, 25957-52-2; **6**, 25906-46-1; **7a**, 25906-47-2; **7b**, 25906-48-3; **7c**, 25898-66-2; **8**, 25906-49-4; **9**, 25906-50-7; **10**, 25906-51-8; **11**, 25906-52-9; **13**, 25898-67-3; 3-methyl-8-nitroindeno[1,2-*c*]pyrazol-4(1*H*)-one azine with acetone, 25906-53-0; 3-methyl-8-nitroindeno[1,2-*c*]pyrazol-4-(1*H*)-one azine with nonanone, 25906-54-1; 5- or 8-hydroxy-3-methylindeno[1,2-*c*]pyrazol-4-(1*H*)-one azine with acetone, 25898-68-4.

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(6) R. Adams, "Organic Reactions," Vol. 1, Wiley, New York, N. Y., 1942, p 163.