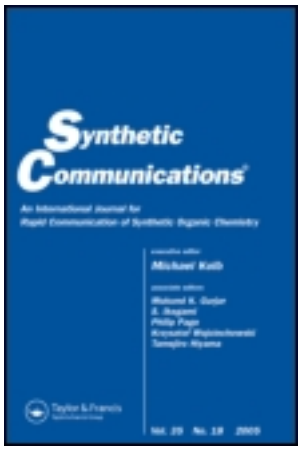

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NOVEL SYNTHESIS OF 4,5-DIHYDROSPIROPYRAZOLE-5,2′-INDANE-1′,3′-DIONES

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ABSTRACT

A novel synthesis of 4,5-dihydrospiropyrazole-5,2′-indane-1′,3′-diones **7a–n**, **8a–c**,**e**,**f** and **12a–f** via the treatment of hydrazonoyl halides **1–3** with 2-arylidene indane-1,3-diones **6a–e** respectively, is reported.

Due to their reactivity and their ease of preparation hydrazonoyl halides[1–8] **1–3** have been used as starting material for the formation of various heterocyclic systems.[9] Now, we report on the reaction of hydrazonoyl halides **1–3** with 2-arylidene indane-1,3-diones **6a–e** prepared as previously described by refluxing of indane-1,3-dione **5** with the aromatic aldehydes in ethanol in the presence of piperidine (Sch. 1). The physical constants obtained for **6a–e** are in agreement with those reported in literature.[10–12]

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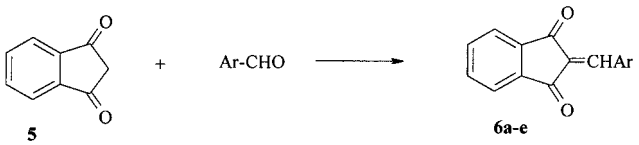
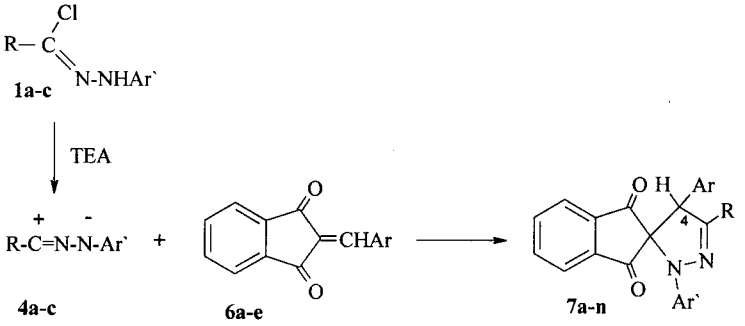


	Ar		Ar
6a	C_6H_5	6d	4-$NO_2C_6H_4$
6b	4-$CH_3C_6H_4$	6e	2-thienyl
6c	4-ClC_6H_4		

Scheme 1.



	R/Ar`/Ar		R/Ar`/Ar
7a	Ph/Ph/Ph	7h	PhCH=CH/Ph/4-ClC_6H_4
7b	Ph/Ph/4-$CH_3C_6H_4$	7i	PhCH=CH/Ph/2-thienyl
7c	Ph/Ph/4-ClC_6H_4	7j	2-thienyl/4-$NO_2C_6H_4$/Ph
7d	Ph/Ph/4-$NO_2C_6H_4$	7k	2-thienyl/4-$NO_2C_6H_4$/4-$CH_3C_6H_4$
7e	Ph/Ph/2-thienyl	7l	2-thienyl/4-$NO_2C_6H_4$/4-ClC_6H_4
7f	PhCH=CH/Ph/Ph	7m	2-thienyl/4-$NO_2C_6H_4$/4-$NO_2C_6H_4$
7g	PhCH=CH/Ph/4-$CH_3C_6H_4$	7n	2-thienyl/4-$NO_2C_6H_4$/2-thienyl

Scheme 2.

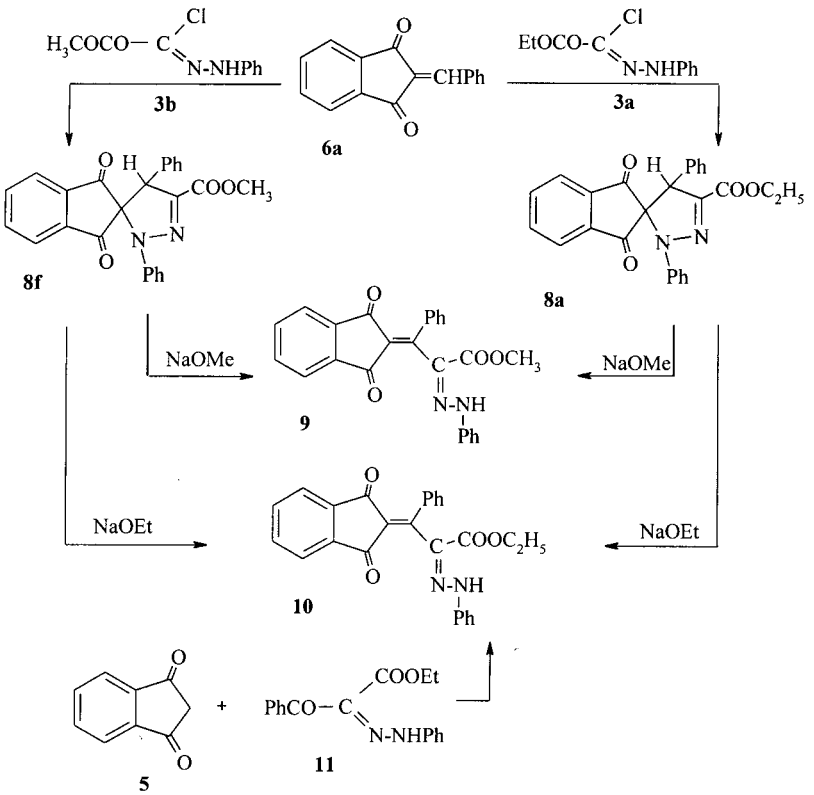
Reaction of 2-arylidene indane-1,3-diones **6a–e** with *C*,*N*-diarylnitrilimines **4a–c**, generated in situ by action of triethylamine on the corresponding *C*,*N*-diarylhydrazonoyl halides **1a–c**, was investigated. Thus, refluxing equimolar amounts of the 2-arylidene indane-1,3-diones **6a–e**, hydrazonoyl halides **1a–c** and triethylamine for 6 h in chloroform gave after work up in each case, only one spirocycloadduct (Sch. 2). All cycloadducts **7** gave satisfactory elemental analyses and the spectral data (IR, ^{1}H NMR, MS) supported the proposed structures. For example,

the ^{1}H NMR spectrum of each compound showed a singlet signal at $\delta \sim 5.20$ ppm assignable to the proton at position 4. This chemical shift agreed with values reported in literature.[13]

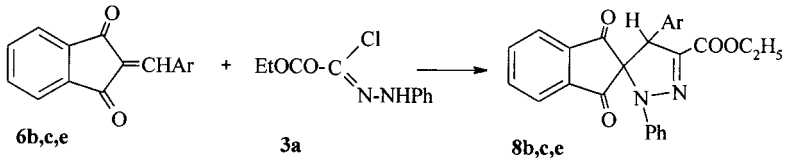
Also, we studied the reaction of 2-arylidene indane-1,3-diones **6a–c**,**e** with α-alkoxycarbonylmethanohydrazonoyl halides **3a**,**b** to investigate the effect of the presence of α-alkoxycarbonyl group on the course of cycloaddition reaction.

Treatment of **6a** with ethyl α-phenylhydrazonochloroacetate **3a** in refluxing chloroform in the presence of triethylamine afforded one product, the spirodione **8a** (Sch. 3). Similarly, reaction of **6a** with methyl α-phenylhydrazonochloroacetate **3b** under similar conditions yielded the respective spirocycloadduct **8f** (Sch. 3).

The structures of the products **8a**,**f** were established on the basis of spectral data and elemental analyses. Also, 2-arylidene indane-1,3-diones



Scheme 3.



	Ar
8b	4-$CH_3C_6H_4$
8c	4-ClC_6H_4
8e	2-thienyl

Scheme 4.

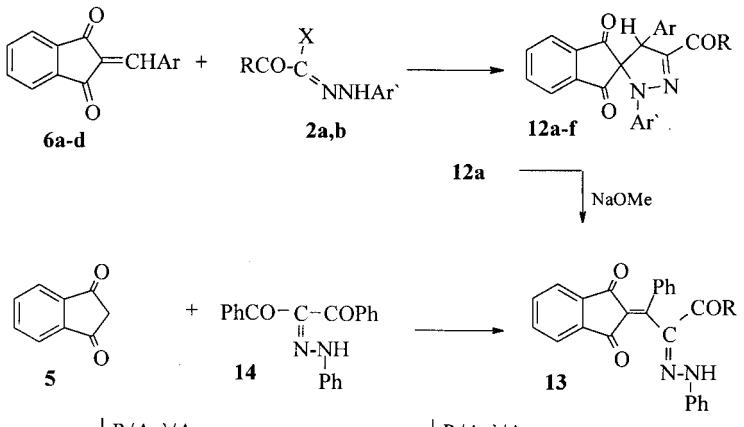
6b,**c**,**e** react with hydrazonoyl halides **3a** to give the corresponding spirocycloadducts **8b**,**c**,**e** respectively (Sch. 4).

Refluxing of either **8a** or **8f** in sodium methoxide in absolute methanol yielded **9**. Similarly **8a** or **8f** gave **10** on boiling in ethanolic sodium ethoxide solution. Further evidence in support of the assigned structure **10** is provided by the alternate synthesis of **10** from indane-1,3-dione **5** and ethyl-3-oxo-2-phenylhydrazono-3-phenylpropanoate[14] **11** (see experimental) (Sch. 3).

Next, the reaction of 2-arylidene indane-1,3-diones **6a–d** with α-keto-hydrazonoyl halides namely α-benzoyl and α-acetylmethano-hydrazonoyl halides **2a** and **2b** respectively, gives the corresponding spirocycloadducts **12a–d**, **12e**,**f** respectively (Sch. 5). The structures of the products were inferred from elemental analyses and spectral data, (see experimental). Treatment of **12a** with sodium methoxide in methanol leads to open chain compound **13**, which also obtained by independent synthesis according to Sch. 5.

EXPERIMENTAL

Melting points were determined on a Gallenkamp electrothermal apparatus and are uncorrected. IR spectra (KBr) were recorded on a Pye Unicam SP-300 IR spectrophotometer and Testscan Shimadzu FTIR 8000 series. ^{1}H NMR spectra were recorded on a Varian Gemini 200 and Varian EM 390 spectrometer using solution in deuterated chloroform and TMS as internal standard. Mass spectra were recorded on a GCMS-QP 1000-EX shimadzu, Japan. Elemental analyses were carried out at the Microanalytical Center, University of Cairo, Giza, Egypt.



	R/Ar`/Ar		R/Ar`/Ar
12a	Ph/Ph/Ph	**12d**	Ph/Ph/4-$NO_2C_6H_4$
12b	Ph/Ph/4-$CH_3C_6H_4$	**12e**	CH_3/ Ph/4-ClC_6H_4
12c	Ph/Ph/4-ClC_6H_4	**12f**	CH_3/ Ph/4-$NO_2C_6H_4$

Scheme 5.

4,5-Dihydrospiropyrazole-5,2′-indane-1′,3′-diones 7a–n, 8a–c,e,f, 12a–f: General procedure: To a mixture of hydrazonoyl halides **1–3** (5 mmol) and 2-arylidene indane-1,3-diones **6a–e** (5 mmol) in chloroform (40 mL), triethylamine (0.7 mL) was added. The reaction mixture was refluxed for 6 h. Then, the solvent was evaporated under reduced pressure and the residue was triturated with methanol (10 mL) and solidified. The crude product was collected and crystallized from suitable solvent. **7a:** m.p. 170°C (acetic acid); 77% yield; ν_{max}/cm^{-1} (KBr); 1746 (C=O), 1714 (C=O); δ_H ($CDCl_3$) 5.2 (s, 1H, pyrazole H-4); 6.7–8.2 (m, 19H, aromatic protons). (Found: C, 81.2; H, 4.5; N, 6.4. $C_{29}H_{20}N_2O_2$ requires C, 81.3; H, 4.7; N, 6.5%). **7b:** m.p. 162°C (ethanol); 87% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1716 (C=O); δ_H ($CDCl_3$) 2.3 (s, 3H, CH_3); 5.2 (s, 1H, pyrazole H-4); 6.6–8.2 (m, 18H, aromatic protons). (Found: C, 81.3; H, 5.0; N, 6.4. $C_{30}H_{22}N_2O_2$ requires C, 81.4; H, 5.0; N, 6.3%). **7c:** m.p. 178°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1748 (C=O), 1714 (C=O); δ_H ($CDCl_3$) 5.2 (s, 1H, pyrazole H-4); 6.8–8.2 (m, 18H, aromatic protons) (Found: C, 75.2; H, 3.9; Cl, 7.8; N, 6.2. $C_{29}H_{19}ClN_2O_2$ requires C, 75.2; H, 4.1; Cl, 7.7; N, 6.1%). **7d:** m.p. 250°C (acetic acid); 77% yield; ν_{max}/cm^{-1} (KBr) 1753 (C=O), 1714 (C=O); δ_H ($CDCl_3$) 5.2 (s, 1H, pyrazole H-4); 6.8–8.2 (m, 18H, aromatic protons); *m/z* 473 (Found: C, 73.4; H, 4.1; N, 8.9. $C_{29}H_{19}N_3O_4$ requires C, 73.6; H, 4.0; N, 8.9%). **7e:** m.p. 205°C (acetic acid); 72% yield; ν_{max}/cm^{-1} (KBr) 1744 (C=O), 1712 (C=O); δ_H ($CDCl_3$) 5.4 (s, 1H, pyrazole H-4);

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6.6–8.2 (m, 17H, aromatic protons); *m/z* 434 (Found: C, 74.4; H, 3.9; N, 6.4; S, 7.4. $C_{27}H_{18}N_2O_2S$ requires C, 74.6; H, 4.2; N, 6.5; S, 7.4%). **7f:** m.p. 155°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1748 (C=O), 1713 (C=O); δ_H ($CDCl_3$) 5.0 (s, 1H, pyrazole H-4); 6.1–8.2 (m, 21H, aromatic protons). (Found: C, 81.7; H, 4.7; N, 6.4. $C_{31}H_{22}N_2O_2$ requires C, 81.9; H, 4.9; N, 6.2%). **7g:** m.p. 211°C (acetic acid); 68% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1714 (C=O); ($CDCl_3$) 2.3 (s, 3H, CH_3); 5.0 (s, 1H, pyrazole H-4); 6.2–8.2 (m, 20H, aromatic protons); *m/z* 468 (Found: C, 82.2; H, 5.0; N, 5.8. $C_{32}H_{24}N_2O_2$ requires C, 82.0; H, 5.1; N, 6.0%). **7h:** m.p. 195°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1713 (C=O); δ_H ($CDCl_3$) 5.0 (s, 1H, pyrazole H-4); 6.1–8.1 (m, 20H, aromatic protons); *m/z* 488 (Found: C, 76.1; H, 4.5; Cl, 7.3; N, 5.6. $C_{31}H_{21}ClN_2O_2$ requires C, 76.1; H, 4.3; Cl, 7.3; N, 5.7%). **7i:** m.p. 195°C (acetic acid); 79% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1715 (C=O); δ_H ($CDCl_3$) 5.3 (s, 1H, pyrazole H-4), 6.3–8.2 (m, 19H, aromatic protons); *m/z* 460 (Found: C, 75.3; H, 4.4; N, 6.2; S, 6.9. $C_{29}H_{20}N_2O_2S$ requires C, 75.6; H, 4.4; N, 6.1; S, 7.0%). **7j:** m.p. 160°C (acetic acid); 77% yield; ν_{max}/cm^{-1} (KBr) 1749 (C=O), 1714 (C=O); δ_H ($CDCl_3$) 5.1 (s, 1H, pyrazole H-4), 6.6–8.7 (m, 16H, aromatic protons). (Found: C, 67.5; H, 3.7; N, 8.7; S, 6.8. $C_{27}H_{17}N_3O_4S$ requires C, 67.6; H, 3.6; N, 8.8; S, 6.7%). **7k:** m.p. 273°C (acetic acid); 83% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1716 (C=O); δ_H ($CDCl_3$) 2.3 (s, 3H, CH_3), 5.2 (s, 1H, pyrazole H-4), 6.7–8.2 (m, 15H, aromatic protons). (Found: C, 68.4; H, 3.9; N, 8.4; S, 6.3. $C_{28}H_{19}N_3O_4S$ requires C, 68.1; H, 3.9; N, 8.5; S, 6.5%). **7l:** m.p. 210°C (acetic acid); 66% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1717 (C=O); δ_H ($CDCl_3$) 5.2 (s, 1H, pyrazole H-4), 6.6–8.2 (m, 15H, aromatic protons); *m/z* 513 (Found: C, 63.1; H, 3.2; Cl, 6.8; N, 8.2; S, 6.3. $C_{27}H_{16}ClN_3O_4S$ requires C, 63.1; H, 3.1; Cl, 6.9; N, 8.2; S, 6.2%). **7m:** m.p. 263°C (acetic acid); 76% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1712 (C=O); δ_H ($CDCl_3$) 5.3 (s, 1H, pyrazole H-4), 6.6–8.3 (m, 15H, aromatic protons); *m/z* 525 (Found: C, 61.8; H, 3.2; N, 10.6; S, 6.2. $C_{27}H_{16}N_4O_6S$ requires C, 61.8; H, 3.1; N, 10.7; S, 6.1%). **7n:** m.p. 200°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1718 (C=O); δ_H ($CDCl_3$) 5.2 (s, 1H, pyrazole H-4), 6.5–8.0 (m, 14H, aromatic protons). (Found: C, 61.6; H, 2.9; N, 8.7; S, 13.1. $C_{25}H_{15}N_3O_4S_2$ requires C, 61.8; H, 3.1; N, 8.7; S, 13.2%). **8a:** m.p. 178°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1719 (C=O); δ_H ($CDCl_3$) 1.2 (t, 3H, CH_3), 4.2 (q, 2H, CH_2), 5.1 (s, 1H, pyrazole H-4), 6.8–8.2 (m, 14H, aromatic protons); *m/z* 424 (Found: C, 73.4; H, 4.5; N, 6.4. $C_{26}H_{20}N_2O_4$ requires C, 73.6; H, 4.8; N, 6.6%). **8b:** m.p. 175°C (acetic acid); 70% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1717 (C=O); δ_H ($CDCl_3$) 1.2 (t, 3H, CH_3), 2.3 (s, 3H, CH_3), 4.2 (q, 2H, CH_2), 5.0 (s, 1H, pyrazole H-4), 6.6–8.2 (m, 13H, aromatic protons). (Found: C, 73.8; H, 5.3; N, 6.3. $C_{27}H_{22}N_2O_4$ requires C, 74.0; H, 5.0;

N, 6.4%). **8c:** m.p. 275°C (acetic acid); 76% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1717 (C=O); δ_H ($CDCl_3$) 1.2 (t, 3H, CH_3), 4.2 (q, 2H, CH_2), 5.0 (s, 1H, pyrazole H-4), 6.8–8.2 (m, 13H, aromatic protons). (Found: C, 68.3; H, 4.3; Cl, 7.8; N, 6.4. $C_{26}H_{19}ClN_2O_4$ requires C, 68.1; H, 4.2; Cl, 7.7; N, 6.1%). **8e:** m.p. 159°C (ethanol); 82% yield; ν_{max}/cm^{-1} (KBr) 1752 (C=O), 1720 (C=O), 1695 (C=O); δ_H ($CDCl_3$) 1.2 (t, 3H, CH_3), 4.3 (q, 2H, CH_2), 5.3 (s, 1H, pyrazole H-4), 6.6–8.2 (m, 12H, aromatic protons). (Found: C, 67.3; H, 4.1; N, 6.4; S, 7.3. $C_{24}H_{18}N_2O_4S$ requires C, 67.0; H, 4.2; N, 6.5; S, 7.5%). **8f:** m.p. 132°C (ethanol); 82% yield; ν_{max}/cm^{-1} (KBr) 1751 (C=O), 1716 (C=O); δ_H ($CDCl_3$) δ 3.7 (s, 3H, CH_3), 5.0 (s, 1H, pyrazole H-4), 6.7–8.1 (m, 14H, aromatic protons). (Found: C, 73.2; H, 4.3; N, 6.6. $C_{25}H_{18}N_2O_4$ requires C, 73.2; H, 4.4; N, 6.8%). **12a:** m.p. 152°C (acetic acid); 75% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1713 (C=O), 1628 (C=O); δ_H ($CDCl_3$) 5.3 (s, 1H, pyrazole H-4), 6.8–8.8 (m, 19H, aromatic protons). (Found: C, 78.7; H, 4.6; N, 6.3. $C_{30}H_{20}N_2O_3$ requires C, 78.9; H, 4.4; N, 6.1%). **12b:** m.p. 180°C (ethanol); 79% yield; ν_{max}/cm^{-1} (KBr) 1746 (C=O), 1712 (C=O); δ_H ($CDCl_3$) 2.3 (s, 3H, CH_3), 5.3 (s, 1H, pyrazole H-4), 6.6–8.4 (m, 18H, aromatic protons). (Found: C, 79.2; H, 4.5; N, 6.2. $C_{31}H_{22}N_2O_3$ requires C, 79.1; H, 4.7; N, 6.0%). **12c:** m.p. 172°C (acetic acid); 76% yield; ν_{max}/cm^{-1} (KBr) 1743 (C=O), 1711 (C=O), 1637 (C=O); δ_H ($CDCl_3$) 5.3 (s, 1H, pyrazole H-4), 6.6–8.4 (m, 18H, aromatic protons). (Found: C, 73.4; H, 3.9; Cl, 7.3; N, 6.0. $C_{30}H_{19}ClN_2O_3$ requires C, 73.4; H, 3.9; Cl, 7.2; N, 5.7%). **12d:** m.p. 160°C (acetic acid); 80% yield; ν_{max}/cm^{-1} (KBr) 1753 (C=O), 1713 (C=O), 1630 (C=O); δ_H ($CDCl_3$) 5.1 (s, 1H, pyrazole H-4), 6.6–8.1 (m, 18H, aromatic protons). (Found: C, 71.8; H 3.7; N, 8.4. $C_{30}H_{19}N_3O_5$ requires C, 71.9; H, 3.8; N, 8.4%). **12e:** m.p. 160°C (acetic acid); 65% yield; ν_{max}/cm^{-1} (KBr) 1750 (C=O), 1711 (C=O); δ_H ($CDCl_3$) 2.6 (s, 3H, CH_3); 5.0 (s, 1H, pyrazole H-4), 6.7–8.2 (m, 13H, aromatic protons). (Found: C, 70.1; H, 4.2; Cl, 8.2; N, 6.4. $C_{25}H_{17}ClN_2O_3$ requires C, 70.0; H, 4.0; Cl, 8.3; N, 6.5%). **12f:** m.p. 177°C (acetic acid); 72% yield; ν_{max}/cm^{-1} (KBr) 1754 (C=O), 1711 (C=O); δ_H ($CDCl_3$) 2.6 (s, 3H, CH_3), 5.1 (s, 1H, pyrazole H-4), 6.9–8.3 (m, 13H, aromatic protons). (Found: C, 68.4; H, 4.2; N, 9.7. $C_{25}H_{17}N_3O_5$ requires C, 68.3; H, 3.9; N, 9.6%).

α[(Phenylhydrazono)methoxycarbonyl]methylbenzylideneindane-1,3-dione 9: To a solution of 4,5-dihydrospiro[1,4-diphenyl-3-ethoxy-carbonyl] pyrazole-5,2′-indane-1′,3′-dione **8a** or 4,5-dihydrospiro[1,4-diphenyl-3-methoxy-carbonyl]pyrazole-5,2′-indane-1′,3′-dione **8f** (1.2 mmol) in methanol (2 mL) was added sodium methoxide (1.2 mmol) and the mixture was boiled for 2 h. The solvent was evaporated and the residue extracted with ether (25 mL) and water (10 mL). The organic layer was dried over anhydrous sodium sulfate and the solvent was evaporated. The residue was recrystallized from acetic acid to give compound **9:** m.p. 235°C (acetic acid); 59% yield;

ν_{max}/cm^{-1} (KBr) 1780 (C=O), 1720 (C=O), 3290 (NH); δ_H ($CDCl_3$) 3.3 (s, 3H, CH_3), 6.5 (s, 1H, NH), 7.0–7.4 (m, 14H, aromatic protons). (Found: C, 73.4; H, 4.3; N, 6.6. $C_{25}H_{18}N_2O_4$ requires C, 73.2; H, 4.4; N, 6.8%).

α-[(Phenylhydrazono)ethoxycarbonyl]methylbenzylideneindane-1,3-dione 10: Method (A). This compound was prepared using the same procedure described for the synthesis of **9** using sodium ethoxide in absolute ethanol in place of sodium methoxide to give compound **10**. **Method (B)**.[14] A mixture of indanedione **5** (1.02 g, 0.7 mmol) and ethylbenzoylacetatephenylhydrazone **11** (2.07 g, 0.7 mmol) was refluxed in ethanol in the presence of sodium ethoxide. The excess solvent was evaporated and the residue was recrystallized from acetic acid to give compound identical in all respects (m.p., mmp, IR, ^{1}H NMR, MS) with **10**. **10:** m.p. 290°C (acetic acid); 60% yield; ν_{max}/cm^{-1} (KBr) 1780 (C=O), 1719 (C=O), 3417 (NH); δ_H ($CDCl_3$) 0.99 (t, 3H, CH_3), 4.1 (q, 2H, CH_2), 6.2 (s, 1H, NH), 6.9–7.3 (m, 14H, aromatic protons); *m*/*z* 424 (Found: C, 73.4; H, 4.7; N, 6.5. $C_{26}H_{20}N_2O_4$ requires C, 73.6; H, 4.8; N, 6.6%).

α[(Phenylhydrazono)benzoyl]methylbenzylideneindane-1,3-dione13: Method (A). This compound was prepared using the same procedure described for the synthesis of **9** using 4,5-dihydrospiro(1,4-diphenyl-3-benzoylpyrazole-5,2′-indane-1′,3′-dione) **12a** in place of **8a** or **8f**. **Method (B)**.[15] Compound **13** was prepared by the same method described for synthesis of **10** using dibenzoylmethanephenylhydrazone **14** in place of **11**. The product was identical in all respects with compound **13** (m.p., mmp, IR, ^{1}H NMR, MS). **13**: m.p. 179°C (acetic acid); 75% yield; ν_{max}/cm^{-1} (KBr) 1773 (C=O), 1655 (C=O), 3296 (NH); δ_H ($CDCl_3$) 6.3 (s, 1H, NH), 6.9–8.9 (m, 19H, aromatic protons). (Found: C, 78.8; H, 4.3; N, 6.2. $C_{30}H_{20}N_2O_3$ requires C, 78.9; H, 4.4; N, 6.1%).

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