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Transformations of Stereoisomeric 2-Chloro-3-R-pentane-1,5-diones in Reaction with Phenylhydrazine

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Abstract—Monophenylhydrazones were prepared from three monochloro derivatives of arylaliphatic 1,5-diketones, and some their transformations were investigated. The monophenylhydrazones formed regiospecifically, and the direction of the reaction was governed by the substituent attached to C^3 atom in the initial chlorodiketones. Formerly unknown products of transformations suffered by monophenylhydrazones obtained were described: 2-amino-1,3-diphenyl-3-(2-phenylindolyl-3)-propan-1-one, 3-benzoyl-2,4,6-triphenyl-2,3,4,5-tetrahydropyridazine, and 3-methyl-1,5-diphenyl-5-phenylazopent-4-en-1-one.

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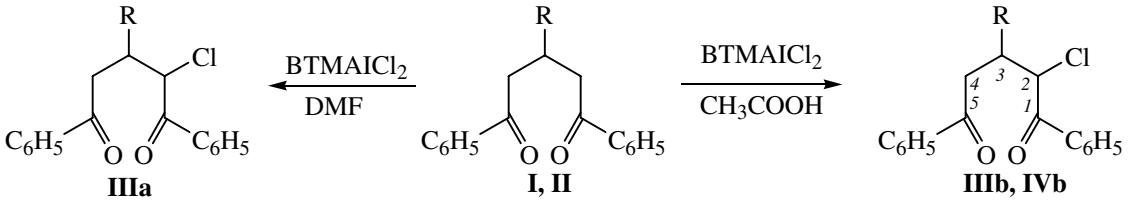
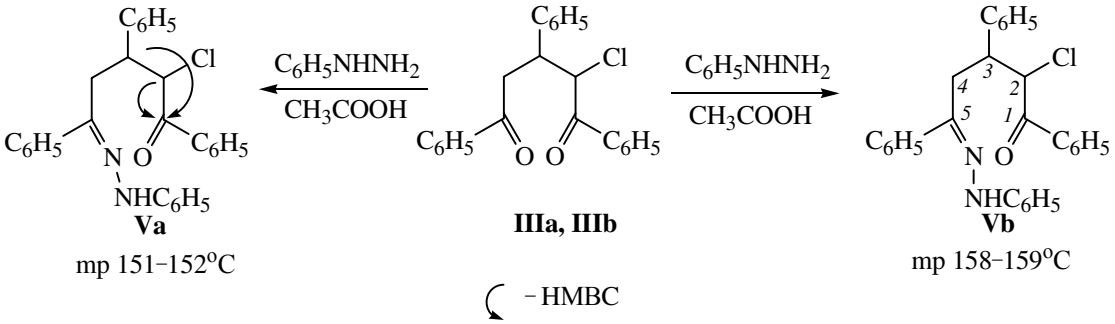
Monochloro derivatives of 1,5-diketone are promising available synthons for preparation of versatile carbocyclic and heterocyclic compounds including also physiologically active substances. These derivatives can be prepared from various 1,5-diketones either by treating with chlorine [1] or with the reagents of tervalent iodine: phenylchloroiodonium chloride ($C_6H_5ICl_2$) and benzyltrimethylammoniumdichloroiodide ($BTMAICl_2$) [2].

Still we believe that the reactivity of both carbonyl groups in these compounds and the range of transformations of monochloro derivatives of arylaliphatic 1,5-diketones are quite insufficiently understood. The aim of this study was investigating reactions of monochloro derivatives of two diketones, 1,3,5-triphenyl-1,5-pentanedione (**I**) and 3-methyl-1,5-diphenyl-1,5-pentanedione (**II**), with phenylhydrazine and subsequent transformations of the corresponding monophenylhydrazones. We obtained monochloro derivative **IIIa** by treating diketone **I** with $BTMAICl_2$ in DMF, and monochloro derivatives **IIIb** and **IVb** by reactions of diketones **I** and **II** respectively with $BTMAICl_2$ in CH_3COOH [2]. Stereoisomeric chloroketone **IVa** we previously obtained in a low yield [2], and it was not used in further study. Compounds **IIIa** and **IIIb** are stereoisomers containing in the molecule two asymmetric atoms C^2 and C^3.

The reaction of γ-haloketones with arylhydrazines in a neutral medium (heating in methanol or ethanol) is known to have resulted formerly in tryptamine derivatives [3]. As follows from the presumed scheme of the reaction [4] the formation of tryptamines from the corresponding 2-chloro-1,5-pentanediones **IIIa**, **IIIb**, and **IVb** inevitably requires in the first stage phenylhydrazones formation at the carbonyl group not linked to the CHCl fragment of the molecule. This condition was actually fulfilled for stereoisomeric monochlorodiketones **IIIa** and **IIIb**. We obtained from these compounds phenylhydrazones **Va** and **Vb** respectively. Their monophenylhydrazone structure was confirmed by IR and mass spectra. The IR spectra



R = C_6H_5 (**I, IIIa, IIIb**), CH_3 (**II, IVa, IVb**).



contain absorption bands at 1682 (C=O), 1602 (C=N), and 3315 (NH) cm^{-1}. In the mass spectra of phenylhydrazones **Va** and **Vb** (electrospray ionization) peaks of pseudomolecular ions appeared at *m/z* 453 $[M + H]^+$.

The assignment of signals in the NMR spectra of compounds synthesized was performed using two-dimensional homo- (COSY) and heteronuclear (HSQC, HMBC) spectroscopy thus permitting unambiguous proof of their structure and identification of the position of the hydrazone fragment at the C^5 atom in compounds **Va** and **Vb**. For instance, the comparison of ^1H and ^{13}C NMR spectra of compounds **IIIa** and **IIIb** and **Va** and **Vb** shows that the CHCl group was conserved in the reaction, and one of the carbonyls transformed into phenylhydrazone. In the ^1H NMR spectra of phenylhydrazones **Va** and **Vb** signals of C<u>H</u>Cl group were observed as doublets which only for hydrazone **Vb** suffered a slight upfield shift compared with the spectrum of the initial chlorodiketone. The coupling constants H^2–H^3 considerably changed in going from the spectrum of chlorodiketone **IIIb** to that of phenylhydrazone **Vb** indicating significant conformational transformations apparently due to introduction into the molecule **Vb** of a bulky phenylhydrazone moiety. The phenylhydrazone character of compounds **Va** and **Vb** was also confirmed by appearance in their ^1H NMR spectra of signals from NH group (8.5 and 8.92 ppm) and also of additional signals from aromatic protons, and in the ^{13}C NMR spectra, of signals belonging to C=N group at 145.4 and 139.0 ppm respectively, and signals of C=O at 193.8 and 194.1 ppm.

The position of the hydrazone moiety in compounds **Va** and **Vb** was successfully established by 2D NMR experiments. For instance, COSY spectra revealed the presence in these compounds of a spin system CH–CH–CH_2, and HSQC experiment revealed a correlation between ^1H and ^{13}C NMR spectra. In HMBC spectrum of phenylhydrazone **Va** cross-peaks were observed for C<u>H</u>Cl (5.58 ppm)/C=O (193.8 ppm) and C<u>H</u>–C_6H_5 (3.86 ppm)/C=O (193.8 ppm). Similarly, in HMBC spectrum of compound **Vb** were found cross-peaks C<u>H</u>Cl (5.54 ppm)/C=O (194.1 ppm) and C<u>H</u>–C_6H_5 (3.96 ppm)/C=O (194.1 ppm).

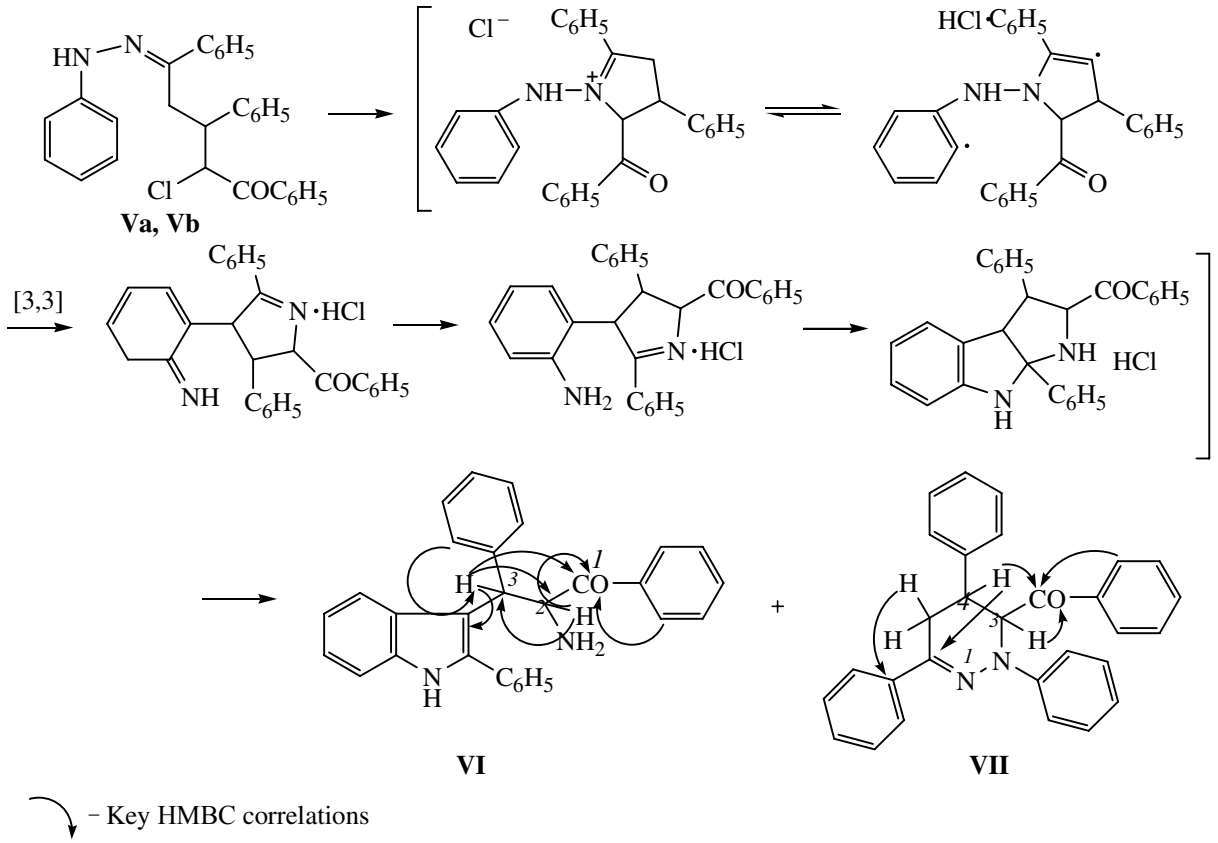
These data unambiguously indicate that the carbonyl group adjacent to CHCl in both phenylhydrazones remains free, and consequently the reaction with phenylhydrazine occurs regiospecifically at a carbonyl far from the CHCl group.

At heating the stereoisomeric hydrazones **Va** and **Vb** in ethanol we obtained the same tryptamine derivative **VI**. This is not in contrast to the presumed scheme [4] of such compounds formation given above in keeping with transformations of compounds **Va** and **Vb**. It follows from the scheme that the reaction involves the CHCl site which undergoes chlorine elimination leading to the loss of the asymmetry distinguishing the initial stereoisomers.

The structure of tryptamine **VI** was confirmed by IR, mass, and NMR spectra, and also by elemental analysis. In the IR spectrum of the compound appeared absorption bands at 1680 (C=O), 3460 (NH of indole ring), 3308, and 3372 (NH_2) cm^{-1}, and in the mass spectrum, a peak of a pseudomolecular ion $[M + H]^+$ at *m/z* 417. ^1H and ^{13}C NMR spectra prove the presence of an indole fragment in compound **VI**. ^1H NMR spectrum contains a singlet signal of NH group at 8.21 ppm, and ^{13}C NMR spectrum, doublet signals at 111.3, 120.3, 121.0, and 122.3 ppm, and also singlets at 111.2, 129.7, 136.4, 137.8, ppm, characteristic of doubly substituted indole ring.

Assignment of signals was carried out by comparison with calculated values and with the use of COSY-45, HSQC, and HMBC experiments. Thus COSY-45 spectrum revealed the nonaromatic CH–CH fragment and the four-proton system of the benzene ring of indole and there three-proton systems of phenyl substituents. The key HMBC correlations, in particular, C^2H (5.5 ppm, d)/C^3 (48.4 ppm, d), CO (202.1 ppm, s), and also C^3H

Scheme.



Va, Vb **VI** **VII**

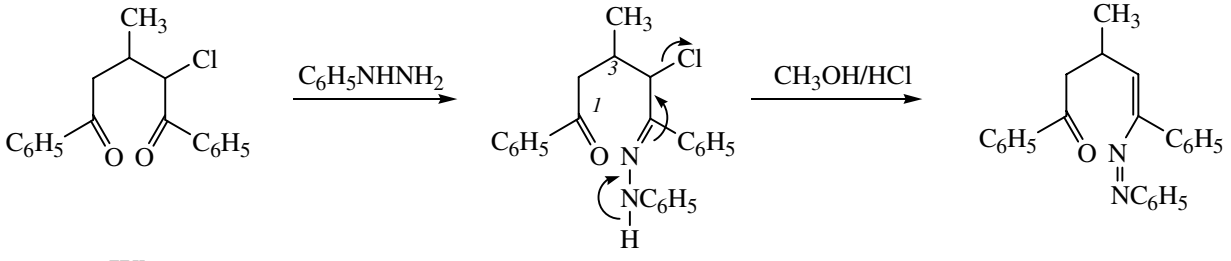
– Key HMBC correlations

(4.87 ppm, d)/C^2 (58.2 ppm, d), CO (202.1 ppm, s) are given on the scheme.

As a side product in reaction of phenylhydrazone **Va** chlorodiketone **IIIa** was isolated, and with phenylhydrazone **Vb**, previously unknown pyridazine **VII**.

A series of substituted tetrahydropyridazines was formerly patented as drugs against osteoporosis, disseminated sclerosis, rheumatoid arthritis, and as phosphodiesterase inhibitors [5]. The structure of pyridazine derivative **VII** was proved by NMR spectroscopy. Spectra COSY-45 revealed a fragment CH–CH–CH_2 and four three-proton spin systems of the phenyl substituents. The neighboring position of the C^{3}H at the carbonyl was proved by HMBC and HSQC experiments, in particular, by cross-peaks C^{3}H (5.71 ppm, t)/CO (196.5 ppm, s) and C^{4}H (3.82 ppm, d.t)/CO (196.5 ppm, s) in HMBC spectrum. The structures both of the major and minor products of hydrazones transformation also proved that in phenylhydrazones **Va** and **Vb** carbon atoms of the CHCl group and of the hydrazone moiety are situated in position *1*,*4*.

Chlorodiketone **IVb** unlike analogous chlorodiketone **IIIa** and **IIIb** in reaction with the phenylhydrazine gave monohydrazone **VIII** at the carbonyl contiguous to CHCl group. The hydrazone at heating in methanol saturated with hydrogen chloride easily transformed into azoolefin



IVb **VIII** **IX**

IX. Similar compounds are known to form readily in 1,4-dehydrohalogenation of α-chloroketones phenylhydrazones [6].

The structure of hydrazone **VIII** got clear after estimation of the structure of the product **IX** of its transformation and analysis of the spectral data. For instance, in the ^{1}H NMR spectrum of compound **VIII** alongside signals of the aromatic protons at 6.9–7.0 ppm and of NH group at 7.80 ppm (s) appear also doublets of a methyl group at 1.20 ppm (*J* 7.0 Hz) and CHCl group at 5.10 ppm (*J* 7.6 Hz), and signals of a methylene group [2.70 d (*J* 3.0 Hz), 2.74 br.s, ppm] and of a methine proton at 2.81 ppm (m). In the corresponding mass spectrum peaks are observed of a pseudomolecular ion of *m/z* 391 $[M + H]^+$ and of ion of 355 $[M + H - HCl]^+$; the IR spectrum contains absorption bands of a carbonyl group at 1682 cm^{-1} and of NH group at 3304 cm^{-1}.

^{1}H NMR spectrum of azoolefin **IX** lacks the signal of NH group and contains doublets from a methyl at 1.21 ppm (*J* 7.0 Hz) and vinyl proton at 5.37 ppm (*J* 1.5 Hz). Methylene group and the adjacent methine proton appear as the following signals, ppm: 2.43 br.d (2H, *J* 6 Hz) and 2.73 m (1H). In the electron-impact mass spectrum of compound **IX** peaks are observed of *m/z* 354 $[M]^+$ and 249 $[M - C_6H_5CO]^+$, and in the IR spectrum, absorption bands at 3056, 1690, 1600, 1560, and 1500 cm^{-1}.

Thus two types of chlorodiketones of a close structure distinguished by a substituent at C^3 atom form monophenylhydrazones regiospecifically either at a remote (R = Ph) or on the contrary at the close to CHCl carbonyl group (R = CH_3). Consequently these monophenylhydrazones can be transformed either into derivatives of indole and pyridazine or into azoolefin. Apparently introducing a chlorine atom into the α-position can increase the reactivity of a carbonyl, but in the presence of a bulky substituent at C^3 atom the approach to this carbonyl is sterically hindered by the substituent and chlorine, and in this event the reaction first of all occurs at the remote carbonyl.

EXPERIMENTAL

^{1}H NMR spectra were registered on spectrometers Bruker DRX-300 and DRX-500 from solutions in deuterochloroform, internal reference TMS. IR spectra were recorded on a spectrophotometer Perkin-Elmer Spectrum BX-II in dichloromethane. HPLC-MS measurements were carried out on Hewlett-Packard Agilent 1100 Series LC/MSD instrument using column Lichro CART CN (4×250 mm, sorbent size 5 μm) at thermostat temperature 40°C in a linear gradient elution mode (30–70% aqueous acetonitrile) at a rate 2% per min. Elution rate was 0.5 ml/min, detecting by electron absorption spectra in the range 200–700 nm, electrospray ionization at atmospheric pressure, mode of positive ions registering, ionization voltage 70 V, ionizing chamber voltage 4 kV, gas-drier flow (nitrogen) 6 l/min, gas-solvent pressure (nitrogen) 50 kg/cm^2. The range of registered masses *m/z* 150–700. Electron-impact mass spectra were measured on LKB 9000S instrument with a direct admission of the sample into the ion source at ionization voltage 70 V.

Melting points of compounds synthesized were measured on a Boëtius heating block. The reaction progress was monitored and the purity of compounds synthesized was checked by TLC on Sorbfil plates, development under UV irradiation or with iodine vapor.

Stereoisomeric 1,3,5-triphenyl-2-chloropentane-1,5-diones (**IIIa** and **IIIb**), and 3-methyl-1,5-diphenyl-2-chloropentane-1,5-dione (**IVb**) were obtained by chlorinating diketones **I** or **II** by $BTMAICl_2$ in acetic acid or DMF respectively in keeping with procedure [2].

1,3,5-Diphenyl-2-chloropentane-1,5-diones phenylhydrazones Va and Vb. To a solution of 0.36 g (1 mmol) of an appropriate chloroketone **IIIa** (R_f 0.25) or **IIIb** (R_f 0.32) in 8 ml of acetic acid prepared by heating at 50–55°C was added dropwise at stirring an equimolar amount of phenylhydrazine (0.11 ml) in 1 ml of ethanol. The precipitate in 3 h phenylhydrazone was filtered off and washed with ethanol.

Phenylhydrazone (**Va**). Yield 0.27 g (60%), mp 151–152°C (from ethanol), R_f 0.45 (hexane–ethyl ether–dichloromethane, 2:0.3:0.1). IR spectrum, ν, cm^{-1}: 1602 (C=N), 1682 (C=O), 3312 (NH). ^{1}H NMR spectrum, δ, ppm: 3.28 d.d (1H, CH_2, *J* 11.2, 14.3 Hz), 3.5 d.d (1H, CH_2, *J* 2.4, 14.0 Hz), 3.86 d.d.d (CH, *J* 2.4, 5.2, 7.6 Hz), 5.58 d (CHCl, *J* 5.0 z), 6.8–8.0 m (20H, Ar), 8.5 s (NH). ^{13}C NMR spectrum, δ, ppm: 30.85 (C^4, CH_2), 42.8 (C^3, CH), 60.8 (C^2, CH), 113.0 (2CH), 119.8 (CH), 125.7 (4CH), 127.4 (CH), 127.9 (4CH), 128.8 (2CH), 128.9 (2CH), 129.1 (2CH), 134.1 (C), 139.0 (C), 140.0 (C), 138.5 (C), 145.4 (C^5), 193.8 (CO). Mass spectrum: *m/z* 453 $[M + H]^+$. Found, %: C 76.80; H 5.30; N 6.10. $C_{29}H_{25}ClN_2O$. Calculated, %: C 76.90; H 5.52; N 6.20.

Phenylhydrazone (Vb). Yield 0.22 g (49%), mp 158–159°C (from ethanol), R_f 0.38. IR spectrum, ν, cm^{-1}: 1602

(C=N), 1680 (C=O), 3315 (NH). ^{1}H NMR spectrum, δ, ppm: 2.90 d.d (1H, CH_2, *J* 3.8, 14.0 Hz), 3.0 d.d (1H, CH_2, *J* 10.1, 14.0 Hz), 3.96 t.d (CH, *J* 3.8, 10.2 Hz), 5.54 d (CHCl, *J* 10.5 Hz), 6.8–8.2 m (20H, Ar), 8.9 s (NH). ^{13}C NMR spectrum, δ, ppm: 32.3 ($C^{4'}$, CH_2), 44.2 ($C^{3'}$, CH), 58.3 ($C^{2'}$, CH), 113.15 (2CH), 119.9 (CH), 125.5 (2CH), 127.2 (CH), 127.75 (2CH), 127.8 (2CH), 128.35 (2CH), 128.7 (2CH), 129.0 (2CH), 129.2 (2CH), 129.4 (2CH), 134.2 (C), 134.25 (CH), 134.6 (CH), 139.0 ($C^{5'}$), 138.5 (C), 134.7 (C), 140.0 (C), 194.1 (CO). Mass spectrum (electrospray ionization): *m/z* 453 $[M + H]^+$. Found, %: C 76.86; H 5.20; N 6.00. $C_{29}H_{25}ClN_2O$. Calculated, %: C 76.90; H 5.52; N 6.20.

2-Amino-1,3-diphenyl-3-(2-phenylindolyl-3)-propan-1-one (VI). In ethanol solution 0.226 g (0.5 mmol) of an appropriate hydrazone **Va** or **Vb** was heated at reflux for 15 h, then the solution was cooled, in reaction with phenylhydrazone **Vb** the precipitate of pyridazine **VII** was filtered off. On distilling off the solvent 10 ml of ethyl ether was added to the residue. The insoluble precipitate of tryptamine **VI** hydrochloride was filtered off. Yield of compound **VI** from hydrazone **Va** 0.1 g (47%), from hydrazone **Vb**, 0.061 g (30%). The hydrochloride was treated with water solution of Na_2CO_3, base **VI** was extracted into ethyl acetate (2×10 ml). The extract was dried over $MgSO_4$, the solvent was distilled off, 0.5 ml of ethanol was added to the residue, the precipitate was filtered off to obtain compound **VI**, mp 170–172°C, R_f 0.22 (hexane–ethyl acetate, 2:1). IR spectrum, ν, cm^{-1}: 1680 (C=O), 3308, 3372 (NH_2), 3460 (NH of indole ring). ^{1}H NMR spectrum, δ, ppm: 4.87 d (C^3H, *J* 8.6 Hz), 5.50 d (C^2H, *J* 8.6 Hz), 7.05 t (CH, *J* 7.4 Hz), 7.14 t (2CH, *J* 7.8 Hz), 7.16 t (CH, *J* 7.0 Hz), 7.25–7.27 m (6CH), 7.35 m (2CH), 7.40 d (CH, *J* 6.8 Hz), 7.41 d (CH, *J* 7.7 Hz), 7.42 d (2CH, *J* 7.0 Hz), 7.69 d (2CH, *J* 7.0 Hz), 7.84 d (CH, *J* 8.0 Hz), 8.21 s (NH). ^{13}C NMR spectrum, δ, ppm: 48.4 (CH), 58.2 (CH), 111.2 (C), 111.3 (CH), 120.3 (CH), 121.0 (CH), 122.3 (CH), 126.3 (CH), 128.25 (5CH), 128.45 (2CH), 128.7 (CH), 128.35 (2CH), 128.7 (2CH), 128.8 (2CH), 129.7 (C), 132.6 (C), 132.7 (CH), 136.1 (C), 136.4 (C), 137.8 (C), 142.3 (C), 202.1 (CO). Mass spectrum: *m/z* 417 $[M + H]^+$. Found, %: C 83.45; H 5.65; N 6.50. $C_{29}H_{24}N_2O$. Calculated, %: C 83.65; H 5.76; N 6.73. Ethyl ether was evaporated from the mother liquor after separating tryptamine **VI** hydrochloride obtained in reaction with phenylhydrazone **Vb** to get an oily substance that slowly crystallized under ethanol. The crystals were filtered off to obtain 0.12 g (53%) of chlorodiketone **IIIb** identified by direct comparison with an authentic sample.

2,4,6-Triphenyl-3-benzoyl-2,3,4,5-tetrahydropyridazine (VII). mp 100–102°C (from ethanol), R_f 0.30 (hexane–ethyl ether–dichloromethane, 2:0.3:0.1). IR spectrum, ν, cm^{-1}: 1690 (CO). ^{1}H NMR spectrum, δ, ppm: 2.74 d.d (1H, CH_2, *J* 7.0, 17.7 Hz), 2.81 d.d (1H, CH_2, *J* 1.6, 7.0 Hz), 3.82 d.t (CH, *J* 1.5, 7.0 Hz), 5.71 t (CH–N, *J* 1.3 Hz), 6.87 t.t (CH, Ar, *J* 1.2, 7.0 Hz), 7.25–7.29 m (10 CH, Ar), 7.37 t.t (2CH, Ar, *J* 1.7, 6.7 Hz), 7.57 t.t (2CH, Ar, *J* 1.7, 7.6 Hz), 7.68 t.t (CH, Ar, *J* 1.8, 7.5 Hz), 7.83 d.t (2CH, Ar, *J* 1.2, 7.1 Hz), 8.11 d.t (2CH, Ar, *J* 1.4, 7.1 Hz). ^{13}C NMR spectrum, δ, ppm: 24.4 (CH_2), 36.1 (CH), 63.1 (CH–N), 114.2 (2CH, Ar), 120.6 (2CH, Ar), 124.9 (2CH, Ar), 126.9 (2CH, Ar), 127.4 (CH, Ar), 127.8 (CH, Ar), 128.2 (2CH, Ar), 128.5 (2CH, Ar), 129.0 (2CH, Ar), 129.1 (2CH, Ar), 129.3 (2CH, Ar), 133.8 (C), 134.0 (CH, Ar), 137.4 (C), 138.5 (C), 142.7 (C), 147.0 (C), 196.5 (CO). Mass spectrum: *m/z* 417 $[M + H]^+$. Found, %: C 83.36; H 5.52; N 6.60. $C_{29}H_{24}N_2O$. Calculated, %: C 83.65; H 5.76; N 6.73.

3-Methyl-1,5-diphenyl-2-chloropentane-1,5-dione phenylhydrazone (VIII). To a solution of 0.5 g (1.7 mmol) of diketone **IVb** in 4 ml of acetic acid was added 0.184 g (1.7 mmol) of phenylhydrazine in 1 ml of ethanol. The solution was stirred by a magnetic stirrer for 1 h. The precipitate of hydrazone **VIII** separated after standing for 10 h at room temperature was filtered off, washed with 3 ml of acetic acid and 3 ml of ethanol. Yield 0.43 g (66%), mp 133–134°C (from ethanol), R_f 0.45 (hexane–ethyl ether–chloroform, 2:0.3:0.1). IR spectrum, ν, cm^{-1}: 1682 cm^{-1} (CO), 3304 (NH). ^{1}H NMR spectrum, δ, ppm: 1.2 d (CH_3, *J* 7.0 Hz), 2.70 d (1H, *J* 3.0 Hz), 2.74 br.C (1H), 2.81 m (1H), 5.1 d (CHCl, *J* 7.6 Hz), 6.9–7.0 m (ArH), 7.8 C (NH). Mass spectrum: *m/z* 391 $[M + H]^+$, 355 $[M + H - HCl]^+$. Found, %: C 73.60; H 5.70; N 7.00. $C_{24}H_{23}ClN_2O$. Calculated, %: C 73.75; H 5.88; N 7.17.

3-Methyl-1,5-diphenyl-5-phenylazopent-4-en-1-one (IX). A solution of 0.9 (2.3 mmol) of hydrazone **VIII** in 10 ml of methanol saturated with hydrogen chloride was heated on a water bath for 7 h and left standing at room temperature for 18 h. The separated viscous substance was isolated, crystallized by adding a mixture ethanol–ethyl ether, 2:1, and recrystallized from ethanol. Yield 0.24 g (30%), yellow crystals, mp 142–144°C, R_f 0.52 (hexane–ethyl ether–chloroform, 2:0.3:0.1). IR spectrum, ν, cm^{-1}: 1500, 1560, 1600, 1690 (CO), 3056. ^{1}H NMR spectrum, δ, ppm: 1.21 d (CH_3, *J* 7.0 Hz), 2.43 br.d (CH_2, *J* 6 Hz), 2.73 m (CH), 5.37 d (CH=, *J* 1.5 Hz). Mass spectrum, *m/z* (electron impact):

354 $[M]^+$, 249. Found, %: C 81.10; H 5.75; N 8.80. $C_{24}H_{21}N_2O$. Calculated, %: C 81.21; H 5.93; N 8.92.

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