Reactions of cyclic enehydrazino ketones with arylidenemalononitriles. Synthesis of 11-aryl-1-oxo-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitriles

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A general and convenient method for the synthesis of 11-aryl-1-oxo-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitriles was elaborated. The method is based on the novel stereoselective rearrangement of fused *N*-arylamino-substituted 1,4-dihydropyridines. The structures of the synthesized compounds were studied using ^{1}H NMR spectroscopy and X-ray diffraction analysis.

Key words: cyclic enehydrazino ketones, arylidenemalononitriles, fused 1,4-dihydropyridines, 11-aryl-1-oxo-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitriles.

Fused heterocyclic systems containing partially hydrogenated pyridine fragments attract permanent attention. In particular, substituted 1,4-dihydropyridines find wide use as medicines for treating cardiovascular diseases.[1] We have previously[2,3] shown that fused *N*-amino-substituted 1,4-dihydropyridines are readily formed by the reactions of cyclic enehydrazino ketones with arylidene derivatives of malononitrile.

It has also been found that *N*-arylamino-substituted 1,4-dihydropyridines **1** undergo a new stereodirected rearrangement to form heterocyclic system **2** containing partially hydrogenated indole and pyridine fragments[3] (Scheme 1). In this work we continued the study of the addition of enehydrazino ketones **3** to arylidenemalononitriles and examined the rearrangement of dihydropyridines **1** that formed to indolo[2,3-*b*]quinolines (quinindolines)* **2**. It is noteworthy that recyclization reactions of various malononitrile derivatives, including arylidenemalononitriles, find wide use in the synthesis of fused heterocyclic systems containing the pyridine ring.[4]

As we have previously shown,[2] short-time heating (5—10 min) of enehydrazino ketones **3** with arylidene derivatives of malononitrile in ethanol is sufficient for the formation of *N*-arylamino-substituted 1,4-dihydropyridines **1**. Subsequent prolonged refluxing (5 h) of products **1** in an ethanolic solution results in their recyclization to quinindolines **2** in 60—80% yields,[3] and refluxing of a mixture of initial substances **3** with arylidenemalononitriles in ethanol for 6 h affords quinindolines **2** in 42—82% yields. Arylidenemalononitriles with both electron-releasing and electron-withdrawing substituents in the aromatic ring enter smoothly into this reaction (Table 1).

The structure of *cis*-9-methyl-1-oxo-11-(4-bromophenyl)-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitrile (**2d**) was established by X-ray diffraction analysis (Fig. 1). It should specially be noted that the H(11) atom (at the C(11) atom) and the cyano group are *trans*-oriented. This indicates that the reaction affords only one isomer of two possible *cis*- or *trans*-isomer **2**. The structure of the products agrees well with the analytical data (Tables 1 and 2). The ^{1}H NMR spectra of products **2** show that the chemical shifts of protons in position 11 depend substantially on the nature of substituents in the benzene ring of the aryl substituent at C(11) (see Table 2), and the chemical shifts of this proton change from 3.90 ppm for electron-releasing substituents to 4.40 ppm for electron-withdrawing substituents.

The found transformation of dihydropyridines **1** into quinindolines **2** is likely related to the known Fischer

* The name *quinindolines* used in *Chemical Abstracts* is not included in the list of trivial names of heterocyclic systems recommended by the rules of the IUPAC nomenclature.

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Table 1. Yields, melting points, conditions of synthesis, and elemental analysis data for compounds **2**

Compound	Substituents			τ*/h	M.p./°C	Yield (%)	Found / Calculated (%)				Molecular formula
	R^1	Ar	R^2				C	H	N	Br	
2a	Me	C_6H_5	H	6	323—325	68	78.56 78.45	5.65 5.76	11.39 11.44	—	$C_{24}H_{21}N_3O$
2b	H	C_6H_5	Me	6	320—322	74	78.24 78.16	5.33 5.42	11.90 11.89	—	$C_{23}H_{19}N_3O$
2c	H	4-BrC_6H_4	H	6	257—259	81	63.30 63.17	3.73 3.86	10.12 10.05	19.01 19.10	$C_{22}H_{16}BrN_3O$
2d	H	4-BrC_6H_4	Me	6	261—263	82	63.79 63.90	4.22 4.20	9.59 9.72	18.55 18.48	$C_{23}H_{18}BrN_3O$
2e	Me	4-BrC_6H_4	Me	6	258—260	61	65.10 65.22	4.96 4.82	9.08 9.13	17.40 17.36	$C_{25}H_{22}BrN_3O$
2f	H	3-$MeOC_6H_4$	H	6	224—226	73	74.69 74.78	5.09 5.18	11.32 11.37	—	$C_{23}H_{19}N_3O_2$
2g	Me	3-$MeOC_6H_4$	Me	6	308—309	42	75.75 75.89	6.26 6.12	10.41 10.21	—	$C_{26}H_{25}N_3O_2$
2h	H	2,5-$(MeO)_2C_6H_3$	Me	6	244—245	73	72.61 72.62	5.60 5.61	10.18 10.16	—	$C_{25}H_{23}N_3O_3$
2i	H	4-$NO_2C_6H_4$	Me	6	283—285	75	69.45 69.34	4.43 4.55	14.25 14.06	—	$C_{23}H_{18}N_4O_3$
2j	H	3-$NO_2C_6H_4$	H	6	263—265	65	68.63 68.74	4.45 4.20	14.67 14.58	—	$C_{22}H_{16}N_4O_3$
2k	H	3,4,5-$(MeO)_3C_6H_4$	Me	2	207—209	51	70.68 70.41	5.55 5.68	9.40 9.47	—	$C_{26}H_{25}N_3O_4$
2l	H	4-$MeOC_6H_4$	Me	2	225—227	55	75.26 75.18	5.61 5.52	11.05 10.96	—	$C_{24}H_{21}N_3O_2$
2k´	H	3,4,5-$(MeO)_3C_6H_4$	Me	20	283—285	42	70.47 70.41	5.73 5.68	9.53 9.47	—	$C_{26}H_{25}N_3O_4$
2l´	H	4-$MeOC_6H_4$	Me	20	309—311	46	75.06 75.18	5.43 5.52	10.84 10.96	—	$C_{24}H_{21}N_3O_2$

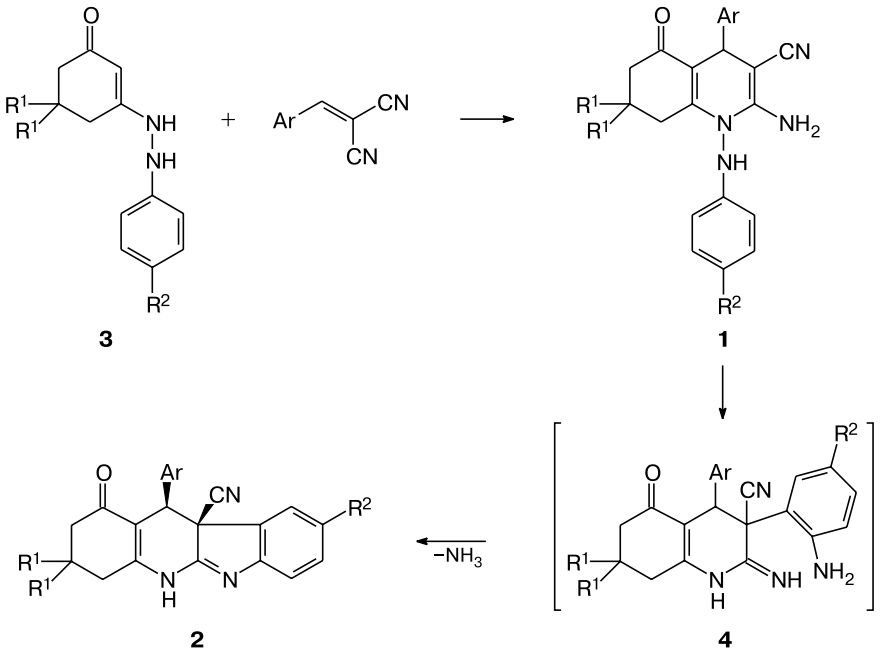
* Duration of the reaction.

synthesis[5] of indoles in which the indole ring is formed with the elimination of an ammonia molecule. The scheme proposed for the formation of compound **2** includes the [3,3] sigmatropic shift similar to the Cope rearrangement, which affords unstable intermediate **4**, and the stage of final intramolecular cyclization with the elimination of an ammonia molecule to form final product **2** (Scheme 1). This transformation scheme accounts for stereoselectivity of the process. Evidently, the second asymmetric center is formed in the stage of [3,3] sigmatropic shift and determined by the configuration of the center in position 4 of the pyridine ring. Further cyclization with the elimination of the ammonia molecule does not result in a loss of overall stereoselectivity of the reaction.

It is worth mentioning, however, that quinindolines **2** obtained from arylidenemalononitriles containing the electron-releasing methoxy substituent in position 4 of the aromatic ring are formed as a mixture of *cis*- and *trans*-isomers **2** and **2´**. The structure of *trans*-9-methyl-1-oxo-11-(3,4,5-trimethoxyphenyl)-2,3,4,5,10b,11-hexahydro-1*H*-quinoline-10b-carbonitrile (**2k´**) was established by X-ray diffraction analysis (Fig. 2). The ^{1}H NMR spectra of the isomers exhibit a substantial difference in chemical shifts of protons in position 4 of the tetrahydropyridine ring. For example, for compound **2k** the chemical shift of this proton is equal to 3.90 ppm, and for isomeric **2k´** it is 5.10 ppm. It is also substantial that the chemical shift of the H(10) proton of the indole ring changes from 6.40 to 7.80 ppm.

Note that in this case the reaction at first occurs stereoselectively: heating of a mixture of the initial substances in ethanol for 2 h affords *cis*-isomer **2** in moderate yield (see Tables 1 and 2). However, the isomerization of **2** to *trans*-isomer **2´** occurs upon prolonged heating (20 h). The transformation of one diastereomer into another includes (Scheme 2), most likely, the formation of zwitterionic intermediate **5**, and the electron-releasing methoxy substituent in position 4 of the aromatic ring

Scheme 1



stabilizes the positively charged part of the intermediate. The tautomerization of **2** to compound **6** can also be the initial stage of the reaction. Opening of the dihydropyridine ring affords intermediate **7**, which is transformed into **2´** through tautomeric form **8** due to intramolecular cyclization. In the absence of the methoxy group, pri-

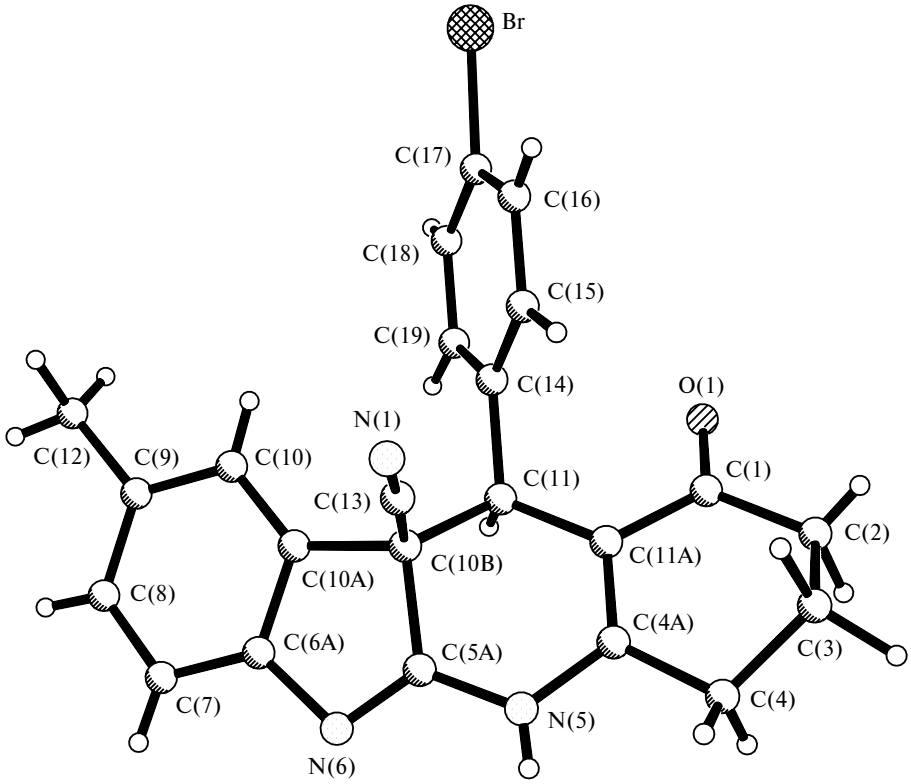


Fig. 1. Structure of compound **2d**.

Table 2. ^{1}H NMR (DMSO-d_6, δ, *J*/Hz) of compounds **2**

Compound	CMe_2	CH_2	CH (s, 1 H)	NH (br.s, 1 H)	Ar	Me (s, 3 H)
2a	1.05 (s, 3 H); 1.25 (s, 3 H)	2.10 (d, 1 H, *J* = 18); 2.35 (d, 1 H, *J* = 18); 2.60—2.70 (m, 2 H)	4.05	11.25	6.35 (d, 1 H, *J* = 8) 7.0 (m, 1 H) 7.30—7.40 (m, 7 H)	—
2b		2.0—2.45 (m, 4 H); 2.60—2.90 (m, 2 H)	4.10	11.35	6.20 (s,1 H); 7.10—7.40 (m, 7 H)	2.20
2c		2.0—2.40 (m, 4 H); 2.60—2.85 (m, 2 H)	4.05	11.30	6.45 (d, 1 H, *J* = 8); 7.05 (m, 1 H); 7.30 (br.d, 2 H); 7.35—7.40 (m, 2 H); 7.60 (d, 2 H, *J* = 8)	—
2d		2.00—2.45 (m, 4 H); 2.60—2.90 (m, 2 H)	4.00	11.25	6.30 (s, 1 H); 7.20—7.35 (m, 4 H); 7.60 (d, 2 H, *J* = 8)	2.20
2e	1.0 (s, 3 H); 1.20 (s, 3 H)	2.10 (d, 1 H, *J* = 18); 2.30 (d, 1 H, *J* = 18); 2.55—2.70 (m, 2 H)	4.10	11.30	6.25 (s, 1 H); 7.10—7.35 (m, 4 H); 7.65 (d, 2 H, *J* = 8)	2.20
2f		1.95—2.40 (m, 4 H); 2.60—2.80 (m, 2 H)	4.10	11.35	3.75 (s, 3 H, OMe); 6.50 (d, 1 H, *J* = 8); 6.75—7.10 (m, 4 H); 7.35—7.50 (m, 3 H)	—
2g	1.05 (s, 3 H); 1.25 (s, 3 H)	2.10 (d, 1 H, *J* = 18); 2.30 (d, 1 H, *J* = 18); 2.55—2.70 (m, 2 H)	4.00	11.25	3.75 (s, 3 H, OMe); 6.28 (s, 1 H); 6.85 (br.s, 1 H); 6.90—7.00 (m, 2 H); 7.20—7.40 (m, 3 H)	2.18
2h		1.95—2.10 (m, 2 H); 2.30—2.40 (m, 2 H); 2.60—2.80 (m, 2 H)	4.00	11.25	3.50 (s, 3 H, OMe); 3.70 (s, 3 H, OMe); 6.30 (s, 1 H); 6.85—7.05 (m, 3 H); 7.18—7.30 (m, 2 H)	2.20
2i		2.00—2.40 (m, 4 H); 2.60—2.80 (m, 2 H)	4.25	11.40	6.30 (s, 1 H); 7.25 (d, 1 H, *J* = 8); 7.35 (d, 1 H, *J* = 8); 7.50—7.70 (br.d, 2 H); 8.30 (d, 2 H, *J* = 8)	2.20
2j		2.00—2.45 (m, 4 H); 2.60—2.90 (m, 2 H)	4.40	11.50	6.50 (d, 1 H, *J* = 8); 7.05 (m, 1 H); 7.40—7.45 (m, 2 H); 7.80—7.85 (m, 2 H); 8.20—8.25 (m, 2 H)	—
2k		1.90—2.30 (m, 4 H); 2.60—2.80 (m, 2 H)	3.90	11.25	6.40 (s, 1 H); 6.60 (br.s, 2 H); 7.23 (d, 1 H, *J* = 8); 7.30 (d, 1 H, *J* = 8)	2.20
2l		1.90—2.30 (m, 4 H); 2.60—2.80 (m, 2 H)	3.95	11.25	6.35 (s, 1 H); 6.90—7.10 (m, 4 H); 7.20—7.30 (m, 2 H)	2.20
2k´		1.90—2.10 (m, 2 H); 2.35—2.45 (m, 2 H); 2.70—2.90 (m, 2 H)	5.10	11.25	6.35 (s, 2 H); 7.00 (m, 2 H); 7.80 (s, 1 H)	2.30
2l´		1.90—2.10 (m, 2 H); 2.35—2.45 (m, 2 H); 2.70—2.90 (m, 2 H)	5.10	11.25	6.60 (d, 2 H, *J* = 8); 6.95 (d, 2 H, *J* = 8); 7.15—7.25 (m, 2 H); 7.70 (s, 1 H)	2.30

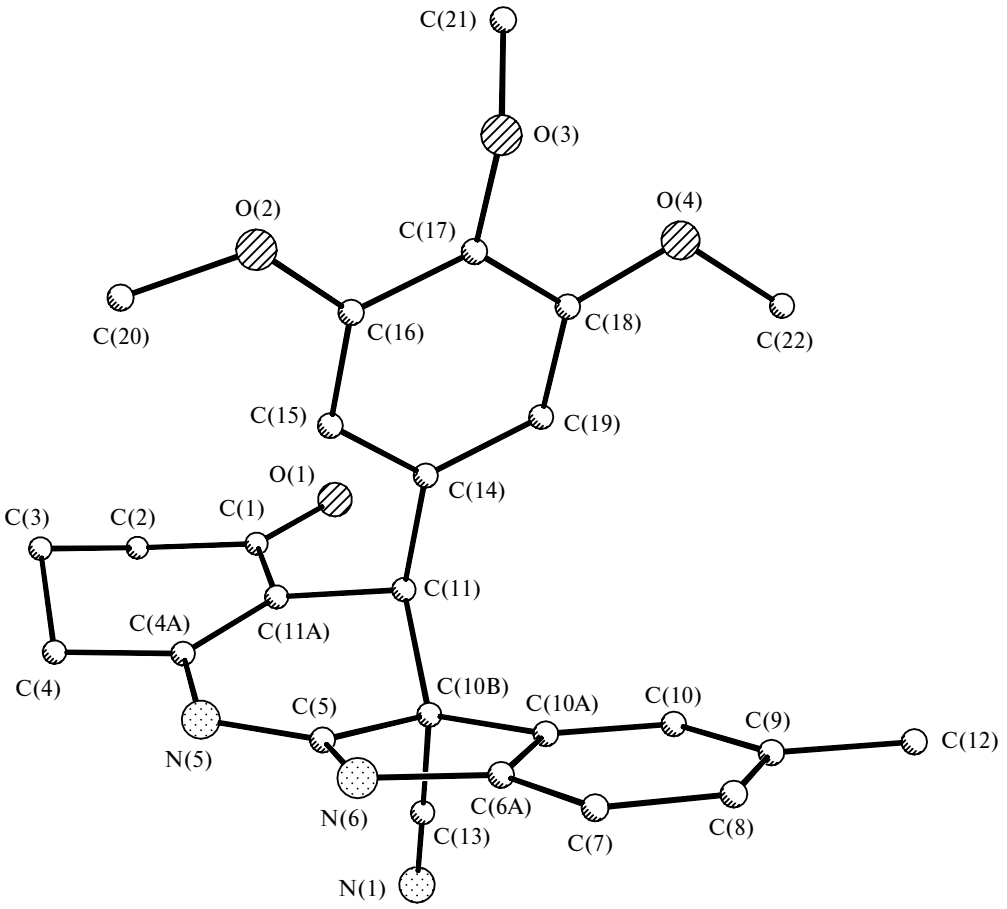


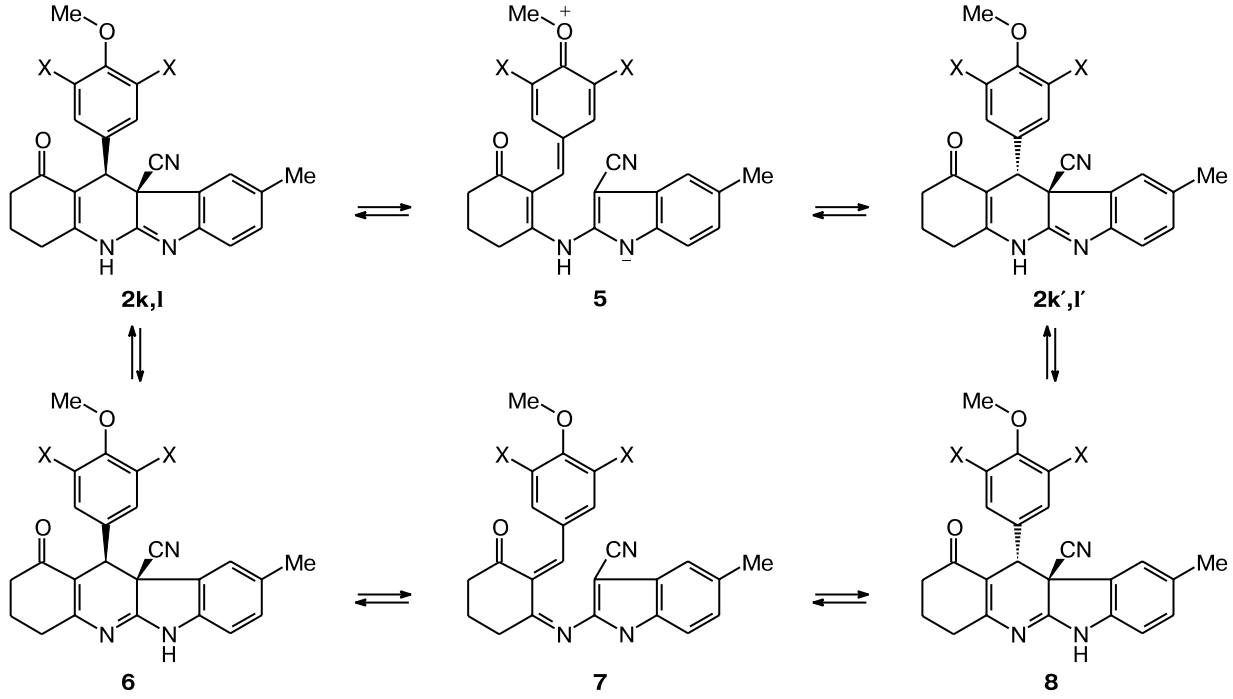
Fig. 2. Structure of compound **2k´**. Hydrogen atoms are not shown.

marily formed *cis*-isomer **2** is kinetically stable and does not isomerize to thermodynamically more stable *trans*-isomer **2´**.

The X-ray diffraction study showed that the substituted six-membered heterocycle in molecule **2d** has a conformation of a distorted half-chair, namely, the C(11) and C(10b) atoms shift from the plane of other four C(5a), N(5), C(4a), and C(11a) atoms (the plane holds with an accuracy of ±0.019 Å) by −0.112 and 0.544 Å, respectively. The conformation of the five-membered heterocycle is a flattened envelope, and the C(10b) atom deviates from the plane drawn through the C(10a), C(6a), N(6), and C(5a) atoms (the deviation of the atoms from the mean plane is ±0.007 Å) by −0.102 Å. The dihedral angle between the planar fragments of the heterocycles equals 159.7° and indicates an insignificant twisting of the molecule, while the indole fragment is planar. The conformation of the cyclohexenone fragment of the molecule is a distorted sofa, in which the C(3) atom shifts from the plane of other five atoms of the ring (the plane holds with an accuracy of ±0.037 Å) by 0.639 Å. The pseudo-equatorial 4-bromophenyl substituent is turned with respect to the planar fragment of the tetrahydropyridine ring by 117.0°. In crystal the intermolecular N(5)—H(N5)...O´ hydrogen bonds (IMHB) (−0.5 + x, 0.5 − y, −0.5 − z) (N(5)...O´ 2.823(4), N(5)—H(N5) 0.75(3), H(N5)...O´ 2.08(3) Å, angle N(5)—H(N5)...O´ 172(2)°) join molecules **2d** to form infinite chains along the c axis.

The conformation of molecule **2k´** differs substantially from that of molecule **2d**. The tetrahydropyridine cycle is substantially flattened compared to molecule **2d**. Its conformation is a slightly distorted sofa: five C(5a), N(5), C(4a), C(11a), and C(11) atoms deviate from their mean plane at most by 0.07 Å, and the C(10b) atom deviates from this plane by 0.657 Å. The indole fragment (C(5a), N(6), C(6a)—C(10b) atoms) is also planar (with an accuracy of ±0.03 Å), and the dihedral angle between the planar fragments is virtually the same as that in **2d** (158.0°). The cyclohexenone fragment has a conformation of a nearly perfect sofa: the C(3) atom deviates from the plane of other five atoms by 0.601 Å. Unlike molecule **2d**, the trimethoxyphenyl substituent in molecule **2k´** is pseudo-axial and turned out with respect to the planar fragment of tetrahydropyridine by 98.3°. The methoxy groups in *meta*-positions are only insignificantly turned out with respect to the benzene ring (by 9.40 and 14.60°), while the *para*-methoxy group is turned out by 104.6°. In crystal of molecule **2k´**, like in structure **2d**,

Scheme 2



X = H (**2k**, **2k**´), X = OMe (**2l**, **2l**´)

N(5)—H(N5)...O´(1) form IMHB (0.5 − *x*, 0.5 + *y*, *z*) with the parameters: N(5)...O´(1) 2.884(4), H(N5)—N(5) 0.92(3), H(N5)...O´(1) 1.98(4) Å, IMHB angle N(5)—H(N5)...O—(1) 165(2)°.

Data on the structure of similar compounds are lacking in the Cambridge Bank of Structural Data (April 2001); therefore, the main geometric parameters of the frameworks of the molecules are presented in Tables 3 and 4. As can be seen from these data, the bond lengths and bond angles for molecules **2d** and **2k**´ virtually coincide and have standard values.[6] However, as said above, the conformations of the frameworks of the molecules differ substantially due to the different orientations of substituents at the C(11) and C(10b) asymmetric centers.

Thus, we studied the reactions of enehydrazino ketones **3** with arylidene derivatives of malononitriles, which afford quinindolines **2** due to the stereoselective thermal rearrangement of compounds **1**. Based on this reaction, we developed the convenient general method for the synthesis of 11-aryl-1-oxo-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitriles (**2**).

Experimental

^{1}H NMR spectra were recorded on Bruker AM-300 (300 MHz) and Bruker WM-250 (250 MHz) instruments in DMSO-d_6. Melting points were measured in a Boetius heating stage and were not corrected. All reaction mixtures were analyzed and purity of isolated products was monitored by TLC

Table 3. Bond lengths (*d*) in molecules **2d** and **2k**´

Bond	*d*/Å	
	Molecule **2d**	Molecule **2k**´
Br(1)—C(17)	1.898(3)	
O—C(1)	1.235(4)	1.232(3)
N(5)—C(5a)	1.361(5)	1.365(3)
N(5)—C(4a)	1.377(4)	1.383(3)
N(6)—C(5a)	1.284(4)	1.283(3)
N(6)—C(6a)	1.420(5)	1.426(3)
N(1)—C(13)	1.141(5)	1.132(3)
C(4a—C(11a)	1.358(4)	1.363(3)
C(4a)—C(4)	1.497(5)	1.502(3)
C(4)—C(3)	1.533(7)	1.514(4)
C(3)—C(2)	1.479(7)	1.510(4)
C(2)—C(1)	1.499(6)	1.511(4)
C(1)—C(11a)	1.469(5)	1.447(5)
C(11a)—C(11)	1.520(5)	1.513(3)
C(11)—C(14)	1.519(4)	1.525(3)
C(11)—C(10b)	1.551(5)	1.550(3)
C(10b)—C(13)	1.474(5)	1.487(3)
C(10b)—C(5a)	1.524(4)	1.529(3)
C(10b)—C(10a)	1.526(5)	1.519(4)
C(10a)—C(10)	1.381(5)	1.374(4)
C(10a)—C(6a)	1.399(4)	1.398(3)
C(10)—C(9)	1.398(5)	1.400(3)
C(9)—C(8)	1.376(5)	1.391(3)
C(9)—C(12)	1.504(6)	1.507(3)
C(8)—C(7)	1.383(6)	1.385(4)
C(7)—C(6a)	1.383(5)	1.383(3)

Table 4. Bond angles (ω) in molecules **2d** and **2k´**

Angle	ω/deg	
	Molecule **2d**	Molecule **2k´**
C(5a)—N(5)—C(4a)	121.2(3)	121.0(2)
C(5a)—N(6)—C(6a)	105.1(3)	105.4(3)
C(11a)—C(4a)—N(5)	121.7(3)	122.7(3)
C(11a)—C(4a)—C(4)	124.3(3)	123.3(3)
N(5)—C(4a)—C(4)	114.0(3)	115.6(3)
C(4a)—C(11a)—C(1)	117.9(3)	120.6(3)
C(4a—C(11a)—C(11)	121.5(3)	120.9(3)
C(1)—C(11a)—C(11)	119.3(3)	118.5(2)
C(14)—C(11)—C(11a)	116.2(3)	113.0(2)
C(14)—C(11)—C(10b)	111.1(3)	112.5(2)
C(11a)—C(11)—C(10b)	108.2(3)	106.7(2)
C(13)—C(10b)—C(5a)	107.1(3)	107.8(2)
C(13)—C(10b)—C(10a)	109.2(3)	110.6(2)
C(5a)—C(10b)—C(10a)	98.2(3)	98.9(2)
C(13)—C(10b)—C(11)	111.4(3)	109.1(2)
C(5a)—C(10b)—C(11)	109.6(3)	108.3(2)
C(10a)—C(10b)—C(11)	120.0(3)	121.1(2)
C(10)—C(10a)—C(6a)	120.8(3)	121.7(2)
C(10)—C(10a)—C(10b)	132.8(3)	132.0(2)
C(6a)—C(10a)—(10b)	106.3(3)	106.3(3)
C(7)—C(6a)—C(10a)	120.5(3)	120.0(2)
C(7)—C(6a)—N(6)	126.3(3)	126.9(2)
C(10a)—C(6a)—N(6)	113.1(3)	113.1(2)
N(6)—C(5a)—N(5)	126.0(3)	126.3(3)
N(6)—C(5a)—C(10b)	116.8(3)	116.1(3)
N(5)—C(5a)—C(10b)	117.2(3)	117.6(2)

using Silufol UV 254 plates with an AcOEt—hexane (3 : 1) mixture as the eluent.

X-ray diffraction study of compounds 2d and 2k´. Colorless single crystals of compounds **2d** and **2k´** appropriate for X-ray diffraction analysis were grown from a solution in acetone. A single crystal of compound **2k´** is the $C_{26}H_{25}N_3O_4 \cdot C_3H_6O$ solvate. The structures were solved by the direct method, which revealed all nonhydrogen atoms, and were refined by the full-matrix least-squares method in the anisotropic approximation for nonhydrogen atoms. Hydrogen atoms were found from the difference electron density syntheses and isotropically refined. The main crystallographic data for these compounds and results of the least-squares method are presented in Table 5. The coordinates of atoms and temperature parameters are deposited at the Cambridge Bank of Structural Data. All calculations were performed using the BSHELXTL PLUS and SHELXTL PLUS+5 program packages.

Synthesis of 11-aryl-1-oxo-2,3,4,5,10b,11-hexahydro-1*H*-indolo[2,3-*b*]quinoline-10b-carbonitriles (2) (general procedure). A solution of enehydrazino ketone **3** (0.001 mol) and the corresponding arylidenemalononitrile (0.001 mol) in ethanol (4 mL) was refluxed for several hours (see Table 1). Then the reaction

Table 5. Main crystallographic parameters for compounds **2d** and **2k´**

Parameter	Molecule **2d**	Molecule **2k´**
Molecular formula	$C_{23}H_{18}BrN_3O$	$C_{26}H_{25}N_3O_4 \cdot C_3H_6O$
Diffractometer	Enraf Nonius CAD 4	Siemens P3/PC
Radiation	MoKα	MoKα
a/Å	9.788(2)	9.762(4)
b/Å	17.733(4)	14.173(7)
c/Å	11.382(2)	38.415(17)
α	90.00	90.00
β	96.24(3)	90.00
γ	90.00	90.00
V/Å^3	1963.9(7)	5315.4(42)
M	432.32	501.57
Space group	$P2_1/n$	$Pbca$
Z	4	8
d_{calc}/g cm^{-3}	1.462	1.254
Scan mode	θ/2θ	θ/2θ
Scan region by θ°	—	2.34—25.05
θ_{max}/deg	28	25.05
Number of independent reflections	5050	4691
Number of reflections with $F^2 \geq 2\sigma(F^2)$	2505	3179
$R_1(F)$	0.055	0.052
w$R_2(F^2)$	0.172	0.115

mixture was cooled, and the precipitate of product **2** was filtered off and washed on the filter with a minor amount of ethanol. The yields and melting points of compounds **2** are presented in Table 1.

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