PLANCHER REARRANGEMENT IN THE REACTION OF 1,3,3-TRIMETHYL-2-CYANOMETHYLENEINDOLINE WITH ortho-PHENYLENEDIAMINE

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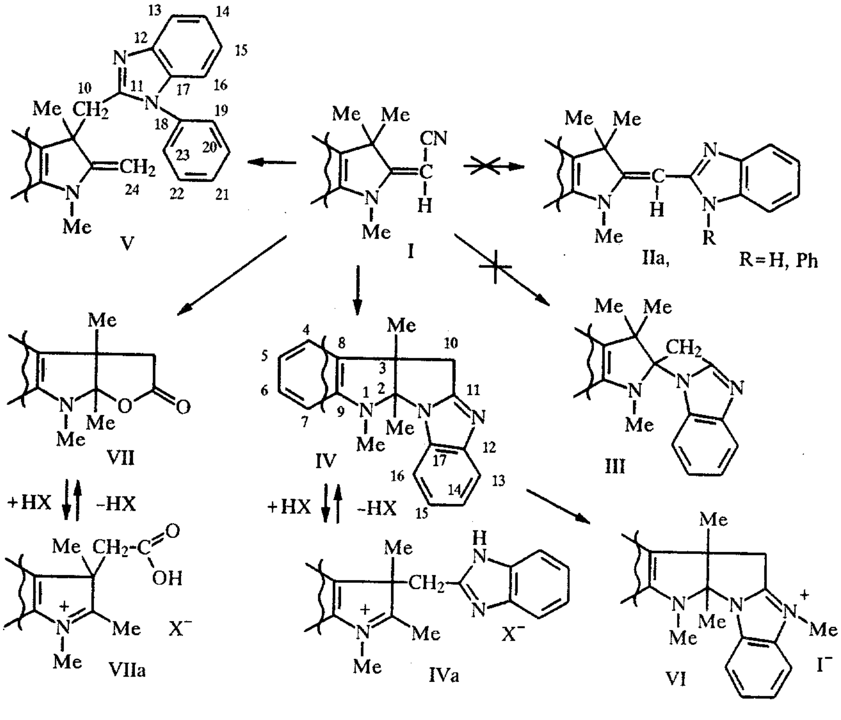
In the reaction of 1,3,3-trimethyl-2-cyanomethyleneindoline with ortho-phenylenediamine in polyphosphoric acid a Plancher rearrangement takes place with the formation of the previously unknown polycyclic system. The structure of the compounds obtained was established by means of x-ray diffraction analysis, ^1H *and* ^{13}C *NMR spectra, and their ring-chain transformations were investigated.*

In continuation of the search for new methods of synthesis of hetaryl-substituted derivatives of 1,3,3-trimethyl-2-methyleneindoline (the Fischer base) [1, 2], we endeavored to synthesize 1,3,3-trimethyl-2-benzimidazolylindolines.

By the condensation of 1,3,3-trimethyl-2-cyanomethyleneindoline I [3] with ortho-phenylenediamine in polyphosphoric acid (PPA) we obtained a pure compound whose elemental analysis corresponded to the presumed structure IIa, but the spectral data contradicted this hypothesis. In the PMR spectrum of the expected 1,3,3-trimethyl-2-(2-benzimidazolyl)methyleneindoline IIa, the presence could be expected of a singlet in the 1.5-1.9 ppm region of two geminal methyl groups at the 3-position and a weak field signal of the methylene group proton at 4.5-5.5 ppm [4, 5]. In reality two singlets are observed of the methyl groups at 1.47 and 1.79 ppm, as well as signals of the CH_2 group (the AB system: $\delta_A = 3.1$; $\delta_B = 3.6$ ppm, $J_{AB} = 17$ Hz) similar to the CH_2 group signals in the spirodihydroisoxazole derivative of 2-methyleneindoline [3].



In the PMR spectra (Table 1) run in different solvents ($CDCl_3$, C_6D_6, DMSO-d_6), the signal of the NH proton is absent, which also indicates that the compound obtained has a cyclic structure. This is also confirmed by the ^{13}C NMR spectrum: the signal of the $C_{(2)}$ carbon atom of the indoline ring is located in the 90.3 ppm region, which corresponds to tetrasubstituted carbon atoms in indolines with a spiro structure of a similar type (90.0-93.6 ppm) [6]. The correctness of the assignment of the $C_{(2)}$ atom is confirmed by recording the ^{13}C NMR spectrum without uncoupling from protons. The signal of the carbon atom in the 157.8 ppm region indicates the presence of a benzimidazole ring in the molecule [7]. The formation of alternative structures

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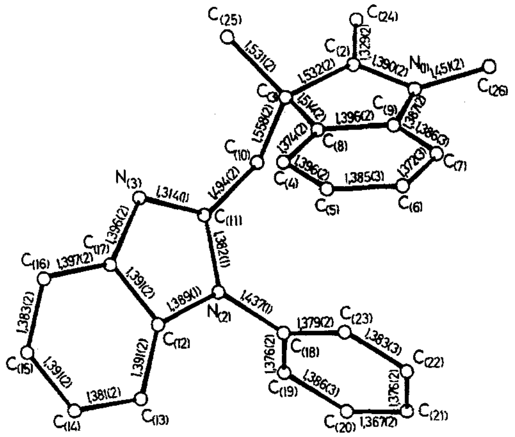

Fig. 1. General view of the molecule of V with numeration of atoms and bond lengths.

TABLE 1. PMR Spectra of Compounds IV-VII

Compound (solvent)	$C_{(3)}CH_3$,	$C_{(2)}CH_3$, [$=CH_2$, (J, Hz)]	NCH_3,	CH_2, d (J, Hz)	7'-H	Aromatic and other protons, ppm
IV ($CDCl_3$)	1,47	1,79	3,22	3,58; 3,14 (17)	6,39	7,54...7,65 (7H, m, Ar)
IVa (CF_3COOD)	1,95	3,08	- 4,18	4,24; 3,80 (14)	7,23	7,30...7,79 (7H, m, Ar)
V ($CDCl_3$)	1,69	[3,65; 3,71; (2,1)]	2,81	2,88; 2,98 (14)		6,44...7,41 (13H, m, Ar)
VI (CD_3CN)	1,55	2,10	3,40	4,00; 3,68 (17)	6,71	3,8 (3H, s, $=N-CH_3$); 6,8...7,9 (7H, m, Ar)
VI (CD_3OD)	1,51	—	3,36	4,06; 3,73 (18)	6,62	3,85 (3H, s, $=N-CH_3$); 6,84 (1H, dd , 5-H); 7,12 (1H, dd , 6-H); 7,36 (1H, d, 4-H); 7,51 (2H, m, 14-H, 15-H); 7,67 (1H, d, 16-H); 7,90 (1H, d, 13-H)
VII ($CDCl_3$)	1,16	1,53	2,81	2,66; 2,86 (17)	6,43	6,71 (1H, dd , 5-H); 6,97...7,11 (2H, m, 4-H, 6-H)

III and IV could be assumed. The formation of the latter structure can be attributed to a Placher rearrangement during the course of the condensation. To confirm the possibility of the realization of such a rearrangement, we carried out the condensation of nitrile I with ortho-aminodiphenylamine, which eliminates any possibility of cyclization at the NH group. In the PMR spectrum of the compound obtained a signal is observed of only one methyl group in the region of 1.65 ppm in the 3-position of the indoline ring, and signals of two CH_2 groups in the form of two doublets each (2.98, 2.88 ppm, J = 14 Hz; 3.78, 3.64 ppm, J = 2.1 Hz). The ^{13}C NMR spectra run with uncoupling from protons and without uncoupling confirmed the presence of two CH_2 groups. These unusual spectral data prompted us to carry out x-ray diffraction investigations.

Figure 1 shows a general view of the molecule with the numeration of atoms and bond lengths. According to RSA, the imidazole ring has the structure usual for benzimidazoles [8]: the ring is absolutely planar, and the bond lengths have intermediate values between the corresponding single and double bonds as the result of the n—π-interaction of the unshared electron pairs (UEP) of the nitrogen atoms with the multiple bonds in the ring. Thus, the $N_{(3)}-C_{(11)}$ bond is delocalized and extended to 1.314 Å in comparison with the usual value of 1.27 Å [9] for this type of a bond, while the neighboring single bonds are correspondingly shortened. The benzene ring planes ($C_{(18)}-C_{(19)}$) and $C_{(3)}C_{(10)}C_{(11)}$ are rotated relative to the plane of the heterocyclic ring by 79.3 and 60.9°. As a result the benzene ring is removed from conjugation with the $N_{(2)}$ atom: the $N_{(2)}-C_{(18)}$ bond length 1.437 Å corresponds to the usual bond length of the type $N_{(sp^2)}-C_{(sp^2)}$ 1.45 Å [9].

The five-membered N-heterocyclic ring in the indoline fragment is substantially nonplanar and has a conformation of a compressed convert. The $C_{(2)}$ atom deviates from the plane of the remaining ring atoms by 0.14 Å, while the bend on the $C_{(3)}...N_{(1)}$ line is not more than 9.6°. The compression of the ring promotes an effective conjugation of UEP of the $N_{(1)}$ atom with the benzene ring and the $C_{(2)}=C_{(24)}$ double bond. A substantial shortening of the $N_{(1)}-C_{(2)}$ and the $N_{(1)}-C_{(9)}$ bond to 1.390(2) and 1.387(2) Å is observed. The $C_{(2)}=C_{(24)}$ double bond has thereby the standard value of 1.329(2) Å (in ethylene

TABLE 2. Valence Angles (ω, deg) in Structure of V

Angle	ω	Angle	ω	Angle	ω
$C_{(2)}N_{(1)}C_{(9)}$	110,8(1)	$C_{(8)}C_{(3)}C_{(25)}$	113,7(1)	$N_{(2)}C_{(11)}C_{(10)}$	121,6(2)
$C_{(2)}N_{(1)}C_{(26)}$	123,5(1)	$C_{(10)}C_{(3)}C_{(25)}$	111,3(1)	$N_{(3)}C_{(11)}C_{(10)}$	125,8(1)
$C_{(9)}N_{(1)}C_{(26)}$	124,5(1)	$C_{(5)}C_{(4)}C_{(8)}$	118,7(2)	$N_{(2)}C_{(12)}C_{(13)}$	131,7(1)
$C_{(11)}N_{(2)}C_{(12)}$	106,6(2)	$C_{(4)}C_{(5)}C_{(6)}$	119,9(2)	$N_{(2)}C_{(12)}C_{(17)}$	105,4(1)
$C_{(11)}N_{(2)}C_{(18)}$	128,3(1)	$C_{(5)}C_{(6)}C_{(7)}$	122,0(2)	$C_{(13)}C_{(12)}C_{(17)}$	122,9(1)
$C_{(12)}N_{(2)}C_{(18)}$	125,2(1)	$C_{(6)}C_{(7)}C_{(9)}$	117,8(2)	$C_{(12)}C_{(13)}C_{(14)}$	116,3(1)
$C_{(11)}N_{(3)}C_{(17)}$	105,0(1)	$C_{(3)}C_{(8)}C_{(4)}$	130,8(1)	$C_{(13)}C_{(14)}C_{(15)}$	121,5(2)
$N_{(1)}C_{(2)}C_{(3)}$	107,7(1)	$C_{(3)}C_{(8)}C_{(9)}$	108,7(1)	$C_{(14)}C_{(15)}C_{(16)}$	121,9(2)
$N_{(1)}C_{(2)}C_{(24)}$	125,9(1)	$C_{(4)}C_{(8)}C_{(9)}$	120,5(1)	$C_{(15)}C_{(16)}C_{(17)}$	117,4(1)
$C_{(3)}C_{(2)}C_{(24)}$	126,4(1)	$N_{(1)}C_{(9)}C_{(7)}$	128,9(1)	$N_{(3)}C_{(17)}C_{(12)}$	110,3(1)
$C_{(2)}C_{(3)}C_{(8)}$	101,7(1)	$N_{(1)}C_{(9)}C_{(8)}$	110,0(1)	$N_{(3)}C_{(17)}C_{(16)}$	129,8(1)
$C_{(2)}C_{(3)}C_{(10)}$	107,1(1)	$C_{(7)}C_{(9)}C_{(8)}$	121,1(2)	$C_{(2)}C_{(17)}C_{(16)}$	119,9(2)
$C_{(2)}C_{(3)}C_{(25)}$	111,8(1)	$C_{(3)}C_{(10)}C_{(11)}$	115,5(1)		
$C_{(8)}C_{(3)}C_{(10)}$	110,7(1)	$N_{(2)}C_{(11)}N_{(3)}$	112,7(1)		

TABLE 3. ^{13}C NMR Spectra of Compounds IV-VII

Compound (solvent)	NCH_3	$C_{(2)}CH_3$ $(=CH_2)$	$C_{(3)}CH_3$	$C_{(3)}$	$C_{(2)}$	$C_{(10)}$	$C_{(7)}$	$C_{(8)}$	$C_{(9)}$	Other carbon atoms
IV ($CDCl_3$)	17,4	22,5	30,4	58,1	90,3	37,1	106,5	134,0	147,8	157,8 ($C_{(11)}$); 110,7; 118,9; 119,6; 121,7; 122,4; 128,7; 131,4
IVa (CF_3COOD)	37,0	—	21,9	59,8	195,9	34,1	116,7	—	—	118,4 ($C_{(13)}$, $C_{(16)}$); 126,6; 131,1; 133,1; 134,1; 134,2; 139,2; 145,1; 148,3
V ($CDCl_3$)	24,3	(73,4)	26,8	46,0	159,4	37,5	103,4	—	—	149,9 ($C_{(11)}$); 103,4; 108,7; 116,7; 117,8; 120,7; 120,9; 121,1; 126,3; 126,4; 126,9; 127,4; 133,1; 134,2; 137,7
VII ($CDCl_3$)	16,3	19,8	26,7	48,9	108,5	40,5	106,1	131,8	146,9	118,7 ($C_{(5)}$); 121,3 ($C_{(4)}$); 127,5 ($C_{(6)}$); 172,3 (C=O)
VII (CD_3OD)	21,5	—	28,7	42,9	176,9	52,1	108,9	135,1	149,9	121,1 ($C_{(5)}$); 124,2 ($C_{(4)}$); 130,2 ($C_{(6)}$)
VIIa (CF_3COOD)	34,1	—	21,7	56,1	195,9	40,9	115,3	138,8	143,1	123,5 ($C_{(5)}$); 131,1 ($C_{(4)}$); 131,6 ($C_{(6)}$); 175,6 (C=O)

the double bond is equal to 1.33 Å [10]). The remaining geometrical parameters of the molecule have standard values.

The established structures of derivative V enabled us to choose structure IV for the compound formed by the condensation of nitrile I with ortho-phenylenediamine. The structure of compound IV is also confirmed by PMR spectra, run in the presence of a lanthanoid shifting reagent (LSR) — $Eu(fod)_3$. The coordination of LSR is realized at the sp^2 hybridized nitrogen atom [11]. When LSR is added, the observed order in the values of the lanthanoid induced shifts (LIS) agrees completely with the structure of IV: 13-H (11.7 ppm) > CH_2 (9.9; 11.2 ppm) > $C_{(2)}$—CH_3 (3.3 ppm) > $C_{(3)}$—CH_3 (2.4 ppm) > N-CH_3 (2.1 ppm).

To prove the structure of compound IV we also studied the deutero exchange. As is known, 2-methyleneindolines and their quaternary salts exchange hydrogen atoms of the methylene and 2-methyl groups for deuterium [12, 13]. In the PMR spectra of indoline IV, run in deuteromethanol, a fairly rapid deutero exchange of the 2-methyl group is observed (in the course of 1 h, the integral peak intensity of the $C_{(2)}$—CH_3 decreased by a factor of almost two).

The PMR spectra of compound IV run in a deuterated trifluoroacetic acid, confirmed the possibility of the existence of this compound in an open form IVa [4]. The general form of the spectrum of CF_3COOD differs little from the spectrum of this compound in $CDCl_3$, but a change is observed in the geminal constant of the spin—spin interaction from 17 to 14 Hz, while in the ^{13}C NMR spectrum in CF_3COOD a characteristic signal of the $C_{(2)}$ carbon atom is observed in the 195.9 ppm region, which also confirms the existence of compound IV in the deuterated trifluoroacetic acid in the acyclic form IVa.

Alkylation of compound IV by methyl iodide results in salt VI. The PMR spectrum of this salt in acetonitrile indicates a cyclic structure of the molecule: the CH_2 group in the form of an AB system with SSCC 17 Hz, signals of four methyl groups in the region of 1.55 ($C_{(3)}—CH_3$); 2.10 ($C_{(2)}—CH_3$); 3.40 ($N—CH_3$); 3.80 ppm ($=N^+—CH_3$). The PMR spectrum of compound VI run in deuteromethanol shows that the deutero exchange proceeds much more rapidly than in compound IV.

Attempts to carry out the cyclization of nitrile I with certain other primary amines [ethylenediamine, 2-hydroxy-2-(p-nitrophenyl)-ethylamine, 2-mercaptobenzimidazole, and 2-aminopyridine] under these conditions turned out to be unsuccessful. In all cases we have isolated only compound VII, which is also obtained by heating nitrile I in PPA, followed by hydrolysis. Compound VII is probably a key compound in the synthesis of compounds IV and V. Compound VII was previously obtained by alkylation of 1,2,3-trimethylindol with bromoacetic acid with subsequent neutralization [12].

In the PMR spectrum of compound VII in $CDCl_3$ signals are observed of three methyl groups: $C_{(3)}—CH_3$ (1.16 ppm), $C_{(2)}—CH_3$ (1.53 ppm); $N—CH_3$ (2.81 ppm), and also signals of the CH_2 group (an AB type spectrum: $\delta_A = 2.86$; $\delta_B = 2.66$ ppm, $J = 17.4$ Hz). The signal of the CH_2 group is observed in the 40.4 ppm region, the signal of the carbonyl group — in the 172.3 ppm region, and the signal of the $C_{(2)}$ carbon atom lies at 108.5 ppm, which agrees with the data for related structures [3, 14]. In the ^{13}C NMR spectrum in CF_3COOD, together with the signal of the carbon atom of the carbonyl group, one more signal appears in the weak field which belongs to the sp^2-hybridized carbon atom (195.9 ppm), and there is also a considerable weak-field shift of the $N—CH_3$ group equal to 17.8 ppm. All this indicates the existence of compound VII in acid media in an acyclic form. It is of interest that lactone VII exists in an acyclic form also in a deuteromethanol solution, as indicated by the data of the ^{13}C NMR spectrum [$C_{(2)}$—176.9 ppm]. In the PMR spectrum of compound VII run in deuteromethanol, signals of the CH_2 group degenerated into a singlet (2.95 ppm) and a rapid deutero exchange of the methylene group is observed.

Thus, the synthesis of compounds IV-VI is possible only when the conditions of the Plancher rearrangement are realized [15].

EXPERIMENTAL

The IR spectra were run on a UR-20 spectrophotometer in KBr tablets, and the ^1H and ^{13}C NMR spectra on a Bruker WP-100 and Varian-200 spectrometer in $CDCl_3$, C_6D_6, CD_3OD, DMSO-d_6, CF_3COOD, using TMS as internal standard. The lanthanoid shifting agent — $Eu(fod)_3$ — a commercial reagent was produced by the Scientific Industrial Association "Reaktiv."

X-Ray Diffraction Investigations. Single crystals of V were monoclinic, at 25°C, $a = 12.928(3)$, $b = 11.850(3)$, $c = 13.916(3)$ Å, $\beta = 108.27(2)°$, $V = 2024(1)$ Å^3, $d_{calc} = 1.16$ g/cm^3, $Z = 4$, sp. gr. $P2_1/n$. The cell parameters and intensities of independent reflections were measured on a CAD-4 automatic 4-disc diffractometer ($\lambda MoK\alpha$, graphite monochromator, ratio of scanning rates: $\theta = 1.2:1.0$, $2\theta \leq 56°$). The structure was interpreted directly using a MULTAN program and refined by a full matrix method of least squares in an anisotropic approximation with respect to 2472 reflections with $F^2 \geq 3\sigma$. All the hydrogen atoms were localized in a different synthesis and at the last stages of refinement were included in the refinement in

TABLE 4. Coordinates of Nonhydrogen Atoms ($\times 10^4$) in the Molecule of Compound V

Atom	x	y	z	Atom	x	y	z
$N_{(1)}$	777(1)	1350(1)	10838(1)	$C_{(13)}$	4882(1)	1876(2)	8104(1)
$N_{(2)}$	3716(1)	1127(1)	9128(1)	$C_{(14)}$	5875(1)	2393(2)	8236(1)
$N_{(3)}$	4877(1)	1473(1)	10676(1)	$C_{(15)}$	6578(1)	2644(2)	9195(2)
$C_{(2)}$	1678(1)	743(1)	11416(1)	$C_{(16)}$	6328(1)	2378(2)	10064(1)
$C_{(3)}$	2697(1)	1305(1)	11296(1)	$C_{(17)}$	5330(1)	1849(1)	9946(1)
$C_{(4)}$	2738(1)	3308(1)	10499(1)	$C_{(18)}$	2758(1)	779(1)	8335(1)
$C_{(5)}$	2097(2)	4189(2)	9963(1)	$C_{(19)}$	2762(1)	-221(2)	7832(1)
$C_{(6)}$	972(2)	4111(2)	9674(2)	$C_{(20)}$	1832(2)	-542(2)	7067(1)
$C_{(7)}$	455(1)	3200(2)	9927(1)	$C_{(21)}$	926(1)	129(2)	6808(1)
$C_{(8)}$	2231(1)	2384(1)	10748(1)	$C_{(22)}$	929(1)	1128(2)	7313(2)
$C_{(9)}$	1097(1)	2338(1)	10478(1)	$C_{(23)}$	1847(1)	1460(1)	8083(1)
$C_{(10)}$	3127(1)	524(1)	10604(1)	$C_{(24)}$	1653(1)	-173(2)	11961(1)
$C_{(11)}$	3923(1)	1054(1)	10162(1)	$C_{(25)}$	3568(1)	1500(2)	12318(1)
$C_{(12)}$	4628(1)	1623(1)	8981(1)	$C_{(26)}$	333(1)	1093(2)	10798(2)

an isotropic approximation. The final divergence factors were $R = 0.036$ and $R_W = 0.051$. All the calculations were carried out on a PDP 11/23+ computer according to an SDP program [16].

The coordinates of the nonhydrogen atoms are given in Table 4; the valence angles are given in Table 2. The results of the elemental analysis of the compounds obtained correspond to the calculated values; the spectral characteristics are given in Tables 1 and 3.

6,6*a*,11*a*-Trihydro-6*a*,11,11*a*-trimethylindolo[2′,3′:1,2]pyrrolo[1,2-*a*]benzimidazole (IV). A 2.5-g portion (13 mmoles) of nitrile I and 1.36 g (13 mmoles) of o-phenylenediamine were added at 100°C to polyphosphoric acid. The mixture was held for 4 h at 190°C, cooled to 100°C, poured into ice water, neutralized with ammonia, and the precipitate was extracted with chloroform. The extract was dried over sodium sulfate, the solvent was evaporated, and the oil obtained was crystallized from isopropanol. Mp 202-204°C. Yield 68%.

1,3-Dimethyl-2-methylene-3-(1-phenyl)-2-benzimidazolylmethylene)indoline (V) was obtained in a similar way as compound IV from nitrile I and o-aminodiphenylamine. The compound was crystallized from isopropanol, mp 146-148°C. Yield 35%.

5-Methyl-6,6*a*,11*a*-trihydro-6*a*,11,11*a*-trimethylindolo[2′,3′:1,2]pyrrolo[1,2-*a*]benzimidazolium Iodide(VI). A 0.15 ml portion (17 mmoles) of methyl iodide was added to 0.5 g (17 mmoles) of compound IV in 10 ml of acetonitrile. The reaction mixture was allowed to stand at room temperature for 24 h, then was heated for 1 h on a boiling water bath, cooled, and 20 ml of ether was added. The precipitate that separated out was filtered off, crystallized from isopropanol, mp 192-194°C. Yield 54%.

2,3,3*a*,8*a*-Tetrahydro-2-oxo-3*a*,8,8*a*-trimethylfuro[2,3-b]indole (VII). A 1.5 g portion of nitrile I in 40 ml of PPA was held at 150°C for 4 h. The reaction mixture was cooled to 100°C, poured into water, neutralized with ammonia, and extracted with chloroform. The extract was dried over sodium sulfate, the solvent was evaporated, and the remaining oil was crystallized from alcohol. MP 127-129°C. IR spectrum (KBr): 1750 cm^{-1} (C=O). Yield 63%.

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