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Liquid-Crystalline 2, 6-Disubstituted Cycloheptimidazoles

B. Krieg [a] & T. Kuhn [a]
[a] Institut für Organische Chemie der Freien Universität, Berlin

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Liquid-Crystalline 2,6-Disubstituted Cycloheptimidazoles

B. KRIEG and T. KUHN

Institut für Organische Chemie der Freien Universität Berlin

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New liquid-crystalline, dichroic 2,6-disubstituted cycloheptimidazoles (1,3-diazaazulenes) have been synthesized and their transition temperatures have been determined by differential scanning calorimetry (DSC). In view of the application of 6-arylazo-substituted cycloheptimidazoles as dyes for electrooptical displays (guest-host-cells), the order parameters of these compounds have been determined.

Keywords: cycloheptimidazoles, order parameter, thermogramms, guest-host dyes

INTRODUCTION

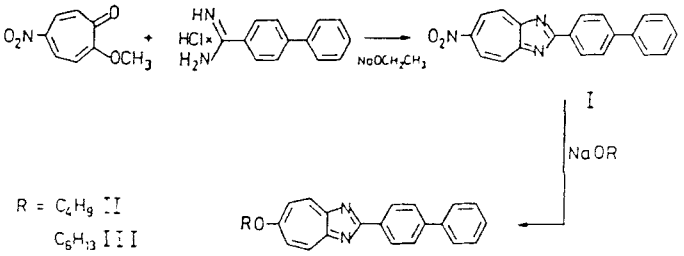
Guest-host cell devices have gained an increased significance as electrooptical displays in recent years because of their wide viewing angle and their coloration.

Continuing the investigations of H. Enzenberg *et al.*,[1] 2,6-disubstituted cycloheptimidazoles were synthesized in view of their application as guest-dyes for the mentioned devices.

RESULTS AND DISCUSSION

Various cycloheptimidazoles were prepared by reaction of 2-methoxy-tropone derivatives with amidines in order to investigate the influence of long chained and "linear" substituents on the liquid-crystalline properties and UV/VIS-absorptions of these derivatives.

For attaching alkyloxy-chains at position 6 of cycloheptimidazole I, the nitro-group was substituted by alkoxides according to a literature procedure.[2]

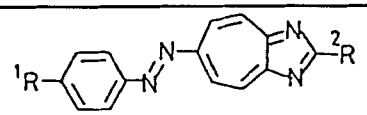


The compound II formed textures of the nematic type and additionally, but only on cooling, a smectic fan texture (Table 2, Figure 1,2). By the cycloheptimidazole III, a smectic A phase with a focal conic, fan-shaped texture followed by a short smectic B phase (lancets) were formed. (Table 2, Figure 1,2,3).

In order to deepen the colour of the cycloheptimidazoles by chromophoric substituents—the UV-absorption maxima of the derivatives II and III did not exceed 400 nm—6-arylazo-cycloheptimidazoles were synthesized.

TABLE I

6-arylazo-cycloheptimidazoles



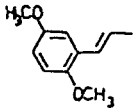
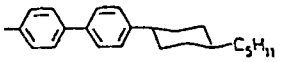
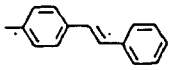
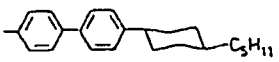
No.	^1R	^2R
IV		
V		
VI		
VII		

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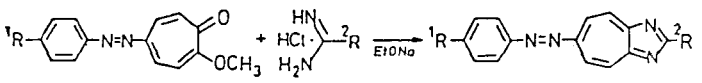
TABLE 2

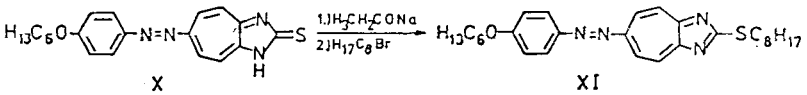
Phase transition temperatures of the cycloheptimidazoles (°C)

No.	C		S_A		S_B		N		I	ΔH (KJ/mol)
II	*	211	*	(207)	—		*	240	*	30.5
III	*	183	*	225	*	228	—		*	
IV	*	263	—		—		*	290	—	31.4
V	*	277	—		—		*	280	—	
VI	*	?	*	237	—		*	300	—	
VII	*	208	*	224	—		—		—	
XI	*	125	*	170	—		—		*	



For the preparation of the cycloheptimidazoles IV, VI and VII 4-cyano-4′-(4-pentylcyclohexyl)-1,1′-biphenyl and all-trans-4-cyano-4′-heptyl-1,1′-bicyclohexane respectively were transformed to the corresponding amidines (VIII and IX) according to the Pinner-method.[3] By using these nitriles, which already have significance as components in liquid-crystalline mixtures,[4] liquid-crystalline properties and high order parameters S of the cycloheptimidazoles should be favored. These expectations were largely fulfilled; nevertheless the observed mesophases of the compounds IV-VII decomposed on heating (Table 2).

However we succeeded in obtaining a thermally stable derivative of the arylazo-substituted species by reaction of the sodium salt of compound X with octyl bromide.



X XI

The compound XI formed a smectic A phase on cooling the isotropic melt: At the transition point I-S_A bâtonnets were observed, which changed with decreasing temperature to form a focal conic, fan texture (Table 2, Figure 1,3).

The order parameters S of the cycloheptimidazoles were determined in guest-host-cells knowing that $S = D - 1/D + 2$. D denotes the dichroic ratio of the absorbances of polarized light parallel and perpendicular to the director n of the liquid crystal mixture.[5] For the meas-

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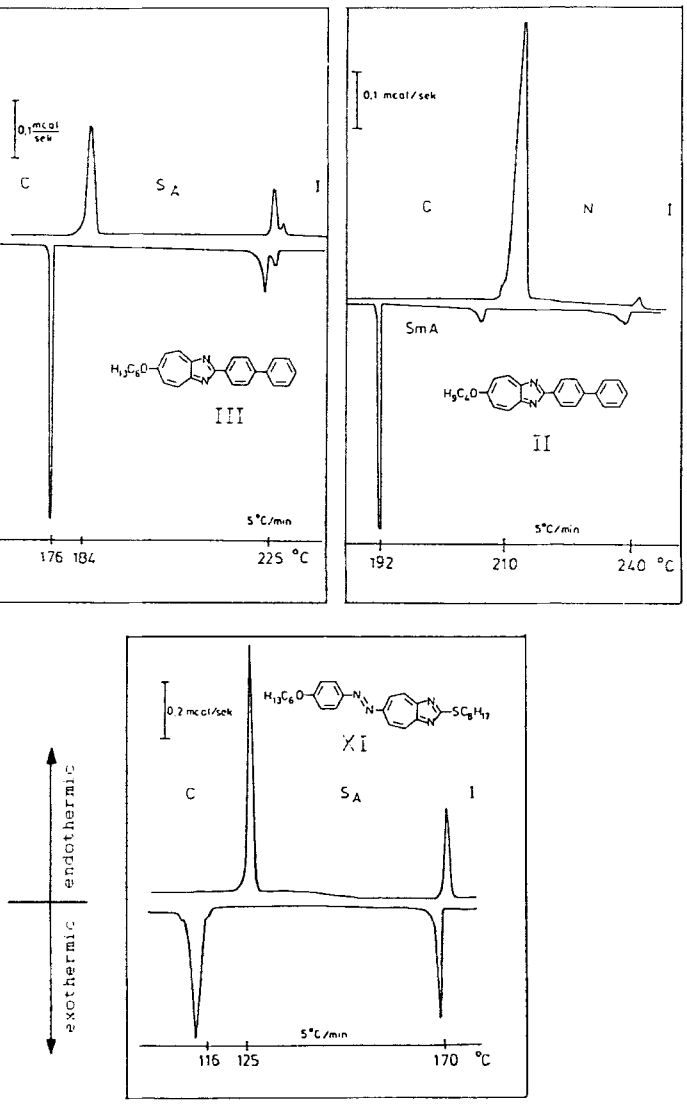

FIGURE 1 Thermograms of the compounds II, III and XI.

urements the liquid-crystalline mixture ZLI-2903 (Merck) has been appplied as host. The measured values for S are listed in Table 3.

The average value 0.84 for S reflects a good parallel alignment of the cycloheptimidazoles to the director n of the liquid-crystalline matrix-mixture.

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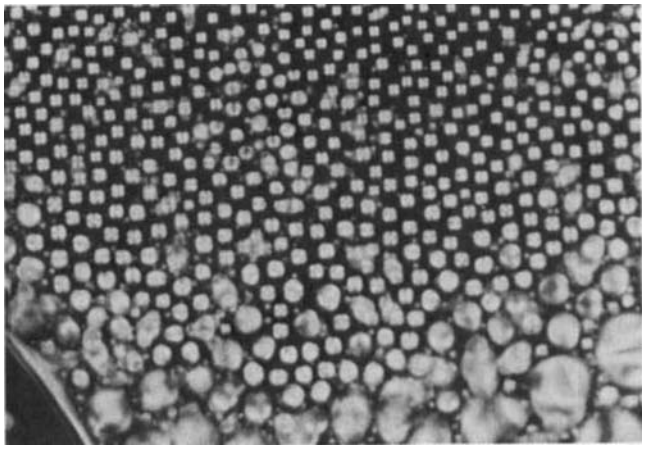
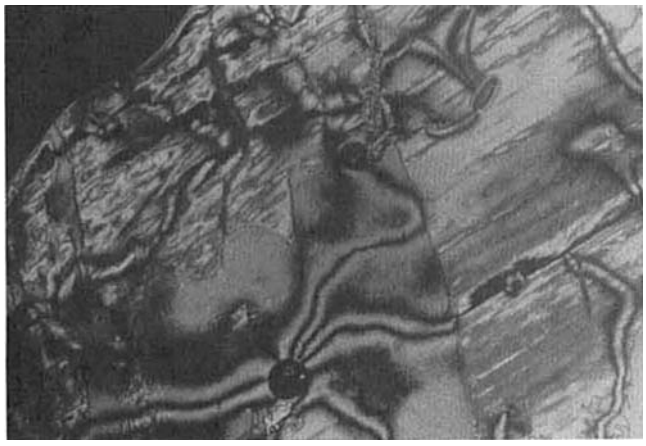
EXPERIMENTAL

Transition temperatures and enthalpies were determined by differential scanning calorimetry using a Perkin-Elmer DSC-2C. The transition temperatures were checked with a polarizing microscope (Zeiss-Universal) equipped with a heating stage. The mesophases of Figures 2,3 were photographed through this microscope with a 126-fold enlargement. The order parameters S were determined by Merck



II 243°C $I \rightarrow N$ nematic droplets



II 240°C N schlieren texture

FIGURE 2

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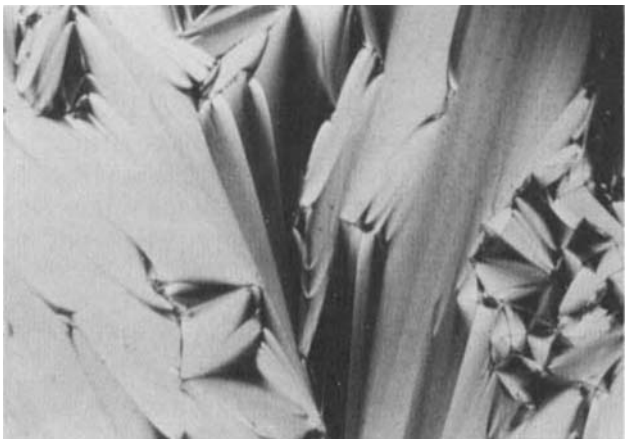
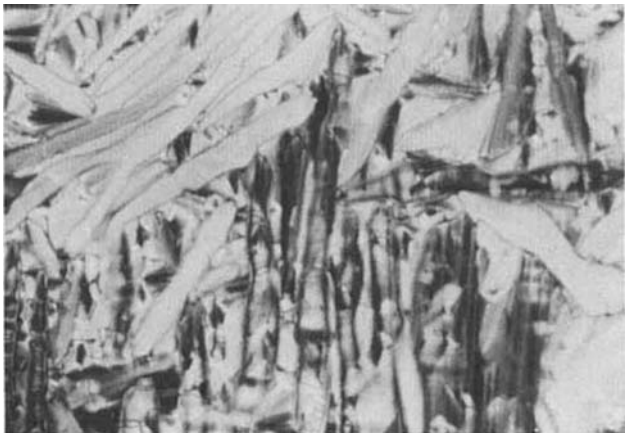


II 208°C S_A fan-shaped texture

FIGURE 2 (*Continued*)

(Darmstadt). The melting points given in the experimental part were measured according to Kofler using a Reichert Thermovar. UV/VIS-spectra were measured using a Perkin-Elmer PE 330 spectrometer and the ^{1}H-NMR-spectra using a Bruker WH-270 spectrometer. The ^{1}H-NMR-data of the cycloheptimidazoles are summarized in Table 4 and the analytical data in Table 5.



III 227°C S_B lancets

FIGURE 3

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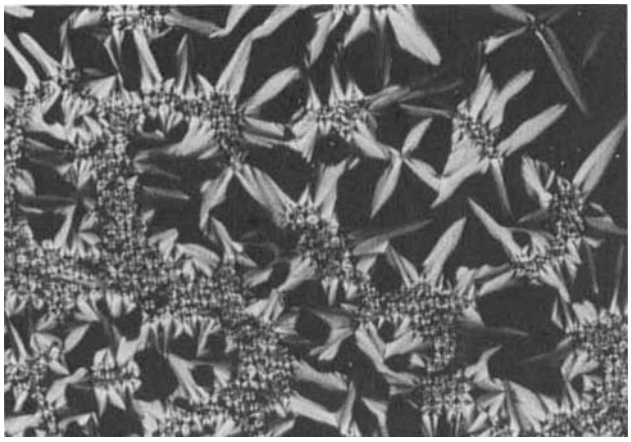
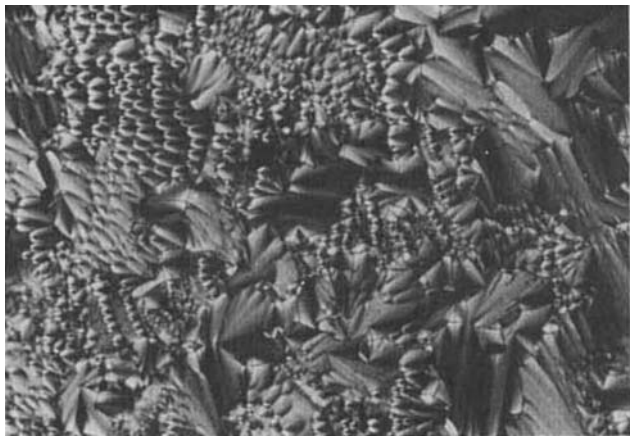


XI 171°C $I \rightarrow S_A$ batonnet texture



XI 170°C S_A fan-shaped, focal conic texture

FIGURE 3 (*Continued*)

TABLE 3

UV-absorption values and order parameters

Nr.	λ_{max}(nm) (CH_2Cl_2)	$E_{1\ cm}$	D	S	λ_{max}(nm) (ZLI-2903)
IV	487	834	17.7	0.85	500
V	464	1166	15.4	0.83	475
VI	459	842	19.1	0.86	468
VII	411	574	14.2	0.82	415

(*E means the absorbance of 1% solutions in dichloromethane*).

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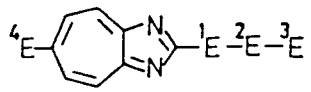
TABLE 4

^{1}H-NMR-data for the 2,6-disubstituted cycloheptimidazoles



No.	solvent	data (δ in ppm)	
I	$DMSOd_6$	seven-membered ring 7.69 (d, J = 12 Hz, 2 H) 6.84 (d, J = 12 Hz, 2 H)	1E = 1,4-phenylene 8.28 (d, J = 9 Hz, 2 H) 7.92 (d, J = 9 Hz, 2 H)
		^{2}E = phenyl 7.71 (d, J = 9 Hz, 2 H) 7.38–7.52 (m, 3 H)	
II	$CDCl_3$	seven-membered ring 8.70 (dd, J = 12a.2 Hz, 2 H) 7.64 (dd, J = 12a.2 Hz, 2 H)	^{1}E = 1,4-phenylene 8.65 (dd, J = 9a.2 Hz, 2 H) 7.87 (dd, J = 9a. 2 Hz, 2 H)
		^{2}E = phenyl 7.72 (dd, J = 9a.2 Hz, 2 H) 7.52–7.38 (m, 3 H)	^{4}E = butyloxy 1.03 (t, J = 7 Hz, 3 H) 1.58 (sex., J = 7 Hz, 2 H) 1.91 (sex., J = 7 Hz, 2 H) 4.24 (t, J = 7 Hz, 2 H)
III	$CDCl_3$	seven-membered ring 8.71 (dd, J = 12a.2 Hz, 2 H) 7.65 (dd, J = 12a.2 Hz, 2 H)	^{1}E = 1,4-phenylene 8.65 (dd, J = 9a. 2 Hz, 2 H) 7.79 (dd, J = 9a.2 Hz, 2 H)
		^{2}E = phenyl 7.71 (dd, J = 9 a.1 Hz, 2 H) 7.36–7.50 (m, 3 H)	^{4}E = hexyloxy 0.94 (t, J = 7 Hz, 3 H) 1.32–1.72 (m, 6 H) 1.96 (q, J = 7 Hz, 2 H) 4.24 (t, J = 7 Hz, 2 H)
IV	$CDCl_3$	seven-membered ring 8.95 (d, J = 11 Hz, 2 H) 8.77 (m, 2 H a.2 H ^{1}E)	^{1}E = 1,4-phenylene 8.77 (m,2 Ha.2 H s.m.r.) 7.82 (d, J = 8 Hz, 2 H)
		^{2}E = 1,4-phenylene 7.67 (m, 2 H a.1 H ^{4}E) 7.34 (d, J = 8 Hz, 2 H)	^{3}E = 4′-pentylcyclohexyl 2.53 (t, J = 14 Hz, 1 H) 1.92 (t, J = 14 Hz, 4 H) 1.00–1.60 (m, 13 H) 0.89 (t, J = 7 Hz, 3 H)
		^{4}E = 4-stilbenylazo 7.74 (d, J = 8 Hz, 2 H) 8.06 (d, J = 8 Hz, 2 H) 7.20 (m, 2 H) 6.88 (s, 2 H) 3.89 (s, 3 H) 3.85 (s, 3 H)	
V	$CDCl_3$	seven-membered ring 8.90 (d, J = 11 Hz, 2 H) 8.72 (d, J = 11 Hz, 2 H)	^{1}E = 1,4-phenylene 8.68 (d, J = 8 Hz, 2 H) 7.73 (d, J = 8 Hz, 2 H)
		^{4}E = 4-tolylazo 7.96 (d, J = 8 Hz, 2 H) 7.45 (d, J = 8 Hz, 2 H) 2.46 (s, 3 H)	^{2}E = 4-stilbenylazo 7.58 (d, J = 8 Hz, 2 H) 7.45–7.18 (m, 5 H + 2 H ^{4}E)

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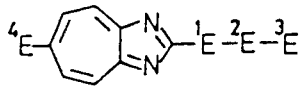
TABLE 4 (*Continued*)

^{1}H-NMR-data for the 2,6-disubstituted cycloheptimidazoles (continuation)



No.	solvent	data (δ in ppm)	
VI	$CDCl_3$	seven-membered ring 8.89 (d, J = 11 Hz, 2 H) 8.70 look at ^{1}E	^{1}E = 1,4-phenylene 8.70 (d + d, J = 11 u.9 Hz, 2 H a.2 H, 7-mem. ring) 7.81 (d, J = 8 Hz, 2 H)
		^{2}E = 1,4-phenylene 7.66 (d, J = 8 Hz, 2 H) 7.34 (d, J = 8 Hz)	^{3}E = 4-pentylcyclohexyl 2.54 (t, J = 14 Hz, 1 H) 1.90 (m, 4 H) 1.10 – 1.60 (m, 13 H, u.6 H hexyloxy)
		^{4}E = 4-hexyloxyphenylazo 8.04 (d, J = 8 Hz, 2 H) 7.06 (d, J = 8 Hz, 2 H) 4.09 (t, J = 7 Hz, 2 H) 0.91 (t a.t, J = 7 Hz, 3 H a.3 H pentyl) 1.90 (m, 2 H)	
VII	$CDCl_3$	seven-membered ring 8.85 (d, J = 11 Hz, 2 H) 8.67 (d, J = 11 Hz, 2 H)	^{1}E = 4′-heptylbicyclohexyl 3.17 (t, J = 14 Hz, 1 H) 2.32 (d, J = 13 Hz, 2 H) 1.70–2.00 (m, 12 H) 1.0–1.60 (m, 17 H) 0.90 (m, 3 H)
		^{4}E = 4-hexyloxyphenylazo	
		8.02 (d, J = 8 Hz, 2 H) 7.04 (d, J = 8 Hz, 2 H) 4.09 (t, J = 7 Hz, 2 H)	1.70–2.00 (m, 2 H) 1.00–1.60 (m, 6 H) 0.90 (m, 3 H)
X	$DMSOd_6$	seven-membered ring 8.48 (d, J = 12 Hz, 2 H) 8.05 (d, J = 12 Hz, 2 H)	^{4}E = 4-hexyloxyphenylazo 7.91 (d, J = 8 Hz, 2 H) 7.26 (m, 2 H) 4.11 (t, J = 7 Hz, 2 H) 1.78 (m, 2 H) 1.10–1.60 (m, 6 H) 0.91 (t, J = 7 Hz, 3 H)
XI	$CDCl_3$	seven-membered ring 8.61 (m, 4 H) ^{4}E = 4-hexyloxyphenylazo 8.02 (d, J = 8 Hz, 2 H) 7.05 (d, J = 8 Hz, 2 H) 4.09 (t, J = 7 Hz, 2 H) 1.87 (m, 2 H) 1.24–1.60 (m, 6 H)	^{4}E = octylthio 3.48 (t, J = 7 Hz, 2 H) 1.87 (m, 2 H) 1.24–1.60 (m, 10 H) 0.93 (m, 3 H + 3 H ^{4}E)

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TABLE 5

Analytical data

	Found %					Calculated %			
Compound	C	H	N	S	Formula	C	H	N	S
II	81.3	6.3	7.9	—	$C_{24}H_{22}N_2O$	81.3	6.3	7.9	—
III	81.7	6.8	7.1	—	$C_{26}H_{26}N_2O$	81.6	6.9	7.3	—
IV	80.4	7.0	8.0	—	$C_{47}H_{48}N_4O_2$	80.5	6.9	8.0	—
V	81.7	5.1	13.4	—	$C_{20}H_{22}N_4$	81.7	5.2	13.1	—
VI	81.1	8.0	8.9	—	$C_{43}H_{50}N_4O$	80.8	7.9	8.8	—
VII	78.3	9.6	9.2	—	$C_{39}H_{56}N_4O$	78.5	9.5	9.4	—
VIII	74.7	8.5	7.2	—	$C_{23}H_{33}N_2Cl$	74.9	8.5	7.3	—
IX	70.4	11.4	8.0	—	$C_{20}H_{39}N_2Cl$	70.0	11.5	8.2	—
XI	70.2	7.9	12.0	7.2	$C_{28}H_{38}N_4O\,S$	70.3	8.0	11.7	6.8
XII	70.9	5.1	7.1	—	$C_{23}H_{20}N_2O_4$	71.1	5.2	7.2	—
XIII	71.5	5.4	6.8	—	$C_{24}H_{22}N_2O_4$	71.6	5.5	6.8	—

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2-[4-(1,1′-Biphenylyl)]-6-nitro-cycloheptimidazole I

To a solution of 2.8 mmol of sodium in 20 ml of absolute ethanol 2.8 mmol of 1,1-biphenyl-4-carboxamidine hydrochloride[6] were added. After stirring for 10 min, 2.8 mmol of 2-methoxy-5-nitro-cycloheptatrien-1-one were added and the mixture was boiled for 2 h. The precipitate in the cooled mixture was filtered off and heated in dioxane. After filtration the concentrated solution afforded brown needles (50%, m.p. 285°C). For the following reactions the product was used as crude material.

UV ($CHCl_3$): λ_{max} (lgϵ) = 269 nm (4.64), 376 (4.01)

2-[4-(1.1′-Biphenylyl)]-6-butyloxy-cycloheptimidazole II

To a solution of 0.6 mmol of sodium in 15 ml of butanol 0.6 mmol of the cycloheptimidazole I was added. After stirring for 2 h at room temperature, the product was filtered off and recrystallized from toluene to give yellow needles (42%).

UV ($CHCl_3$): λ_{max} (lgϵ)

= 282 nm (4.50), 297.5 (4.42), 380 (4.58), 400,5 (4.59).

2-[4-(1.1′-Biphenylyl)]-6-hexyloxy-cycloheptimidazole III

To a solution of 1.1 mmol of sodium in 15 ml of hexanol 1.1 mmol of the cycloheptimidazole I was added. After stirring for 2 h at 50°C and cooling, the product was filtered off and recrystallized from toluene to give yellow needles (25%).

UV ($CHCl_3$): λ_{max} (lgϵ)

= 270 nm (4.50), 276 (4.50), 294 (4.43), 387 (4.56), 400 (4.61).

6-(4-[2-(2,5-Dimethoxyphenyl)-ethenyl]-phenylazo)-2-(4-[4′-(trans-4-pentylcyclohexyl)]-1,1′-biphenylyl)-cycloheptimidazole IV

1.0 mmol of the 2-methoxytropone derivative XIII, 1.0 mmol of the amidine hydrochloride VIII and 1.0 mmol of sodium were reacted in the same manner as described above for the preparation of I. The

precipitate was filtered off and heated in toluene. After filtration the concentrated solution afforded violet needles, which were recrystallized from toluene/chloroform (33%).

6-[4-Methylphenyl)azo]-2-[4-(2-phenyl-ethenyl)-phenyl]-cycloheptimidazole V

1.6 mmol of 2-methoxy-5-[4-methylphenyl)azo]-tropone,[1] 1.6 mmol of sodium and 1.6 mmol of 4-stilbene-carboxamidine hydrochloride[6] were reacted as described for the preparation of IV. The product was recrystallized from toluene to give red scales (50%).

6-[(4-Hexyloxyphenyl)azo]-2-(4-[4'-(trans-4-pentyl-cycloxhexyl)]-1-,1'-biphenylyl)-cycloheptimidazole VI

1.0 mmol of 5-[(4-hexyloxyphenyl)azo]-2-methoxy-tropone,[7] 1.0 mmol of sodium and 1.0 mmol of the amidine hydrochloride derivative VIII were reacted as described for the preparation of IV. Recrystallization of the product from chloroform afforded red crystals (49%).

6-[4-Hexyloxyphenyl)azo]-2-[all-trans-4-(4'-heptyl-1,1'-bicyclohexyl)]-cycloheptimidazole VII

1.5 mmol of 5-[(4-hexyloxyphenyl)azo]-2-methoxy-tropone,[7] 1.5 mmol of sodium and 1.5 mmol of the amidine hydrochloride derivative IX were reacted as described for the preparation of IV. The product was recrystallized from toluene to give red crystals (39%).

4-[4'-(Trans-4-pentylcyclohexyl)-1,1'-biphenylyl]-carboxamidine hydrochloride VIII

According to the Pinner-method,[3] 12 mmol of 4-cyano-4'-(trans-4-pentylcyclohexyl)-1,1'-biphenyl (Merck) were transformed to the corresponding imidic ester by reaction with 24 mmol of abs. ethanol and hydrogen chloride in dioxane (44%, m.p. 295°C). This product was dissolved in a saturated solution of ammonia and absolute ethanol to give the corresponding amidine hydrochloride (83%, m.p. 215°C).

4-(All-trans-4'-heptyl-1,1'-bicyclohexane)-carboxamidine hydrochloride (IX)

35 mmol of 4-cyano-all-trans-4'-heptyl-1,1'-bicyclohexane (Merck) were transformed to the corresponding imidic ester as described above for VIII (59%, m.p. 220–224°C). After reaction with ammonia the amidine hydrochloride was obtained (76%, m.p. 325–333°C).

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6-[(4-Hexyloxyphenyl)azo]-2(1H)-cycloheptimidazole-thione X

1.7 mmol of 5-[(4-hexyloxyphenyl)azo]-2-methoxy-tropone,[7] 1.7 mmol of sodium and 1.7 mmol of thiourea were reacted according to a prescribed procedure.[8] Brown crystals were obtained (71%, m.p. 244–247°C) and used in the following reaction as crude material.

6-[(4-Hexyloxyphenyl)azo]-2-octylthio-cycloheptimidazole XI

1.4 mmol of the cycloheptimidazole derivative X, 1.4 mmol of sodium and 1.4 mmol of octyl bromide were reacted according to a prescribed procedure.[8] The product was dissolved in toluene/ethanol 10/2 and passed through a column of silica gel. The crystalline residue obtained on evaporation of the eluant, was recrystallized from methanol/ether to give red cyrstals (15%).

UV ($CHCl_3$): λ_{max} (lgϵ) = 259 nm (4.46), 323 (3.95), 451 (4.60)

5-{4-[2-(2,5-Dimethoxyphenyl)-ethenyl]phenylazo}-2-hydroxy-2,4,6-cycloheptatrien-1-one (XII)

16 mmol of 4-amino-2′,5′-dimethoxystilbene (FLUKA) were diazotised at 0°C with an equimolar amount of sodium nitrite in a mixture of 25 ml of water and 14 ml of 25% hydrochloric acid. The resulting solution was poured at 0°C into a solution of 16 mmol of tropolone in 12 ml of pyridine. After stirring for 30 min, the precipitate was collected by filtration, washed with water and dried over phosphorus pentoxide. The product was recrystallized from toluene, giving brown crystals (65%, m.p. 171–173°C).

5-{4-[2-(2,5-Dimethoxyphenyl)-ethenyl]phenylazo}-2-methoxy-2,4,6-cycloheptatrien-1-one (XIII)

The methylation of XII (11 mmol) with diazomethane according to a prescribed method[7] gave reddish brown needles, which were recrystallized from chloroform (95%, m.p. 160–161°C).

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