



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>4</sup> :</b>  <b>C09K 19/42, 19/34</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 86/ 02375</b>  <b>(43) International Publication Date:</b> 24 April 1986 (24.04.86)
<b>(21) International Application Number:</b> PCT/EP85/00529 <b>(22) International Filing Date:</b> 10 October 1985 (10.10.85)  <b>(31) Priority Application Number:</b> 84112484.5 (EP) <b>(32) Priority Date:</b> 17 October 1984 (17.10.84) <b>(33) Priority Countries:</b> DE, et al.  <b>(71) Applicants (for all designated States except US):</b> MERCK PATENT GESELLSCHAFT MIT BESCHRÄNKTER HAFTUNG[DE/DE]; Frankfurter Strasse 250, D-6100 Darmstadt (DE). TOSHIBA CORPORATION [JP/JP]; 8, Shinsugita-cho, Isogo-ku, Yokohama 235 (JP).  <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only) :</b> MATSUMOTO, Shoichi [JP/JP]; TOMII, Hitoshi [JP/JP]; Toshiba Corp., 8, Shinsugita-cho, Isogo-ku, Yokohama 235 (JP). SCHEUBLE, Bernhard [DE/DE]; Am Grenzweg 18, D-6146 Alsbach (DE). WEBER, Georg [DE/DE]; Wilhelm-Leuschner-Str. 38, D-6106 Erzhausen (DE). SAGE, Ian, Charles [GB/GB]; BDH Chemicals, Ltd., Broom Road, Poole BH12 4NN (GB).		<b>(74) Common Representative:</b> MERCK PATENT GESELLSCHAFT MIT BESCHRÄNKTER HAFTUNG; Frankfurter Str. 250, D-6100 Darmstadt (DE).  <b>(81) Designated States:</b> DE, GB, US.  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> LIQUID CRYSTAL COMPOSITION  <b>(57) Abstract</b>  Liquid crystal compositions containing at least one compound of formula: R-Dio-Ph-CN wherein R is alkyl of 2 to 5 carbon atoms, Dio is trans-1,3-dioxane-2,5-diyl and Ph is 1,4-phenylene, and at least one compound of each of the following groups A to D: A) R <sup>1</sup> -Cy-Ph-R <sup>2</sup> , B) R <sup>3</sup> -Cy-Ph-Ph-R <sup>4</sup> , C) R <sup>5</sup> -Cy-Ph-CN, D) R <sup>6</sup> -Cy-COO-Ph-O-R <sup>7</sup> and, in addition in a total amount of 10 to 26% by weight at least five compounds of formula E R <sup>8</sup> -Pyr-Ph-O-R <sup>9</sup> or at least one compound of each of the following groups F to K: F) R <sup>10</sup> -Ph-COO-Ph-R <sup>11</sup> , G) R <sup>12</sup> -Cy-COO-Ph-Ph-CN, H) R <sup>13</sup> -Ph-Ph-COO-Ph-Ph-CN, J) R <sup>14</sup> -Ph-COO-Ph-COO-Ph-R <sup>15</sup> , K) R <sup>16</sup> -Ph-Ph-COO-Ph-R <sup>17</sup> result in surprisingly steep electrooptical characteristics and surprisingly low viewing-angle dependence of the contrast at simultaneously acceptable threshold voltages for commercially available drivers and multiplex ratios up to 1:100 when used in matrix displays.		

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## Liquid Crystal Composition

The invention relates to liquid crystal compositions suitable for use in electro—optical matrix displays of the twisted nematic cell type, and to an electro-optical matrix display using the liquid crystal composition mentioned above.

There is still a great demand for liquid crystal compositions (LC compositions) suitable for matrix displays having an electrooptical characteristic (contrast/voltage curve) as steep as possible and simultaneously a low threshold voltage. In such matrix displays, each "display point" of an electrode grid can be selectively triggered by applying a voltage to a first electrode layer, consisting of a multiplicity of horizontal rows of conductors, and to another electrode layer, consisting of a multiplicity of vertical rows of conductors (multiplex drive). The higher the multiplex ratio and the lower the maximum voltage of the available drivers, the lower the threshold voltage of the LC composition has to be. A disadvantage of these matrix display elements is the partial activation of display points in the immediate vicinity of a triggered display point, which partial activation is called "crosstalk" and reduces the contrast of the display in an undesirable manner. The steeper the electro-optical characteristic of the LC composition used in a display element of this type, the less such crosstalk is then observed. As a rule, the steepness of the electro-optical characteristic of a LC composition is given as the ratio of the control voltages  $V$  which must be applied to a given display element in order to obtain 10% of the maximum contrast ( $V_{10}$ ) and 50% of the maximum contrast ( $V_{50}$ ).

The steepness of the electro-optical characteristic is the greater, the smaller the ratio

$$\gamma = \frac{V_{50} - V_{10}}{V_{10}} \cdot 100 \text{ [\%]}$$

Till today the steepness of the electro-optical characteristic can only be determined by experiment, because all theoretical predictions ended up to now in rather wrong results.

Besides of a decrease of the contrast with increasing multiplex ratio, there is also a strongly increasing viewing-angle dependence of the contrast. This dependence is defined as

$$\beta = \frac{V_{10,0,25} - V_{10,40,25}}{\frac{1}{2} (V_{10,0,25} + V_{10,40,25})} \cdot 100 \text{ [\%]}$$

wherein  $V_{x,y,z}$  is the applied control voltage at x % contrast, viewing angle y and temperature z °C.

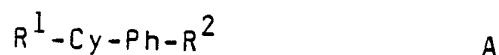
The smaller  $\beta$ , the better is the optical performance of the matrix display.

It has now been found that LC compositions according to the invention result in surprisingly steep electro-optical characteristics and surprisingly low viewing-angle dependence of the contrast at simultaneously acceptable threshold voltages for commercially available drivers and multiplex ratios up to 1 : 100 when used in matrix displays.

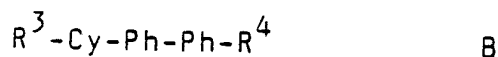
The invention thus relates to a liquid crystal composition, characterized in that it contains at least one compound of the formula I



- 5 wherein R is alkyl of 2 to 5 carbon atoms, Dio is trans-1,3-dioxane-2,5-diyl and Ph is 1,4-phenylene, and at least one compound of each of the following groups A to D:



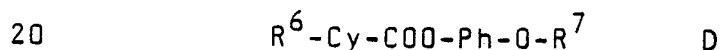
- 10 wherein  $\text{R}^1$  and  $\text{R}^2$  are each alkyl of 3 to 5 carbon atoms, Cy is trans-1,4-cyclohexylene and Ph is 1,4-phenylene,



- 15 wherein  $\text{R}^3$  and  $\text{R}^4$  are each alkyl of 2 to 5 carbon atoms and Cy and Ph have the meanings indicated above,

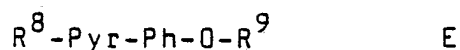


wherein  $\text{R}^5$  is alkyl of 2 to 5 carbon atoms and Cy and Ph have the meanings indicated above,



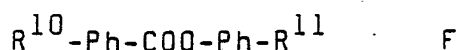
- wherein  $\text{R}^6$  and  $\text{R}^7$  are each alkyl of up to 5 carbon atoms and Cy and Ph have the meanings indicated above, and, in addition in a total amount of 10 to 26% by weight at least five compounds of the  
25 formula E

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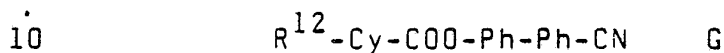


wherein  $R^8$  and  $R^9$  are each alkyl of 5 to 11 carbon atoms, Pyr is pyrimidine-2,5-diyl and Ph has the meaning indicated above,

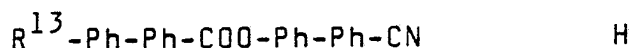
5 or at least one compound of each of the following groups F to K:



wherein  $R^{10}$  and  $R^{11}$  are each alkyl of 1 to 7 carbon atoms, Ph has the meaning indicated above,



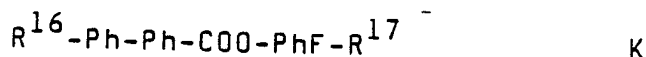
wherein  $R^{12}$  is alkyl of 1 to 3 carbon atoms, and Cy and Ph have the meaning indicated above.



wherein  $R^{13}$  is alkyl of 6 to 8 carbon atoms  
15 and Ph has the meaning indicated above.



wherein  $R^{14}$  and  $R^{15}$  are each alkyl of 1 to 5 carbon atoms and Ph has the meaning indicated above,



wherein  $R^{16}$  and  $R^{17}$  are each alkyl of 5 to 7 carbon atoms, PhF is fluorinated 1,4-phenylene and Ph has the meaning indicated above,

- 5 and to an electro-optical display device, characterized that it contains a liquid crystal composition according to Claim 1.

The LC compositions according to the invention preferably contain at least two compounds of the formula I  
10 preferably in a total amount of 10 to 25, preferably 11,5 to 20 % by weight.

The LC compositions according to the first aspect of the invention preferably contain only one compound of the formula A preferably in an amount of 5 to 15% by  
15 weight, preferably at least two compounds of the formula B preferably in an total amount of 15 to 25% by weight and preferably only one compound of the formula C preferably in an amount of 5 to 10 % by weight. These LC compositions according to the invention  
20 preferably contain at least three compounds of the formula D preferably in a total amount of 10 to 26 % by weight.

$R^6$  preferably is straight chain alkyl of 3 to 5 carbon atoms.  $R^7$  preferably is methyl.

- 25 These LC compositions according to the invention preferably contain five compounds of the formula E preferably in a total amount of 20 to 25 % by weight.

$R^8$  preferably is n-hexyl.

The LC compositions according to the second aspect of the invention preferably contain at least six compounds of the formula F preferably in a total amount of 50 to 65 % by weight and preferably only one  
5 compound of the formula G preferably in an amount of 5 to 15 % by weight,  $R^{12}$  is preferably ethyl.

These LC compositions according to the invention preferably contain only one compound of the formula H preferably in an amount of 1 to 5 % by weight.  $R^{13}$   
10 is preferably n-heptyl.

These LC compositions according to the invention preferably contain only one compound of the formula J preferably in an amount of 3 to 7 % by weight.

$R^{14}$  and  $R^{15}$  are preferably n-propyl.

15 These LC compositions according to the invention furthermore preferably contain only one compound of the formula K preferably in an amount of 7 to 11 % by weight.

$R^{16}$  is preferably n-heptyl and  $R^{17}$  is preferably  
20 n-pentyl and PhF is preferably 2-fluoro-1,4-phenylene.

The preparation of the LC compositions according to the invention is carried out in a manner which is conventional per se. The single components are known from the prior art. As a rule, the desired quantity of  
25 the components used in a smaller quantity is dissolved in the component(s) representing the main constituent, advantageously at an elevated temperature. If the temperature selected here is above the clearing point of the main constituent, the completeness of the  
30 solution process can be observed with particular ease.



It is also possible, however, to mix solutions of the components in a suitable organic solvent, for example acetone, chloroform or methanol, and, after thorough mixing, to remove the solvent again, for example by  
5 distillation under reduced pressure. Of course, it is necessary in this procedure to take care that no impurities or undesired doping substances are introduced by the solvent.

The examples which follow are intended to explain the  
10 invention without restricting it. Parts or percentage figures denote parts by weight or per cent by weight respectively.

#### Example A

A LC composition consisting of

- |    |      |  |
|----|------|--|
| 15 | 7 %  | 2-p-cyanophenyl-5-propyl-1,3-dioxane,                              |
|    | 7 %  | 2-p-cyanophenyl-5-butyl-1,3-dioxane,                               |
|    | 6 %  | 2-p-cyanophenyl-5-pentyl-1,3-dioxane,                              |
|    | 8 %  | p-trans-4-propylcyclohexyl-benzonitrile,                           |
|    | 12 % | 4-ethyl-4'-(trans-4-propylcyclohexyl)-biphenyl,                    |
| 20 | 11 % | 4-ethyl-4'-(trans-4-pentylcyclohexyl)-biphenyl,                    |
|    | 3 %  | 2-p-pentoxyphenyl-5-hexylpyrimidine,                               |
|    | 3 %  | 2-p-hexoxyphenyl-5-hexylpyrimidine,                                |
|    | 4 %  | 2-p-heptoxyphenyl-5-hexylpyrimidine,                               |
|    | 6 %  | 2-nonoxyphenyl-5-hexylpyrimidine,                                  |
| 25 | 6 %  | 2-p-undecoxyphenyl-5-hexylpyrimidine,                              |
|    | 7 %  | trans-4-propylcyclohexancarboxylic acid-(p-methoxyphenylester),    |
|    | 5 %  | trans-4-butylcyclohexancarboxylic acid-(p-methoxyphenylester),     |
| 30 | 4 %  | trans-4-pentylcyclohexancarboxylic acid-(p-methoxyphenylester) and |
|    | 11 % | trans-1-p-propylphenyl-4-pentylcyclohexane                         |

has a clear point of 61°C and a viscosity of 31 mPa·s at  
35 20°C.

## Example B

A LC composition consisting of

	8.2 %	4-(5-ethyl-1,3-dioxane-2-yl)benzonitril
	3.3 %	4-(5-butyl-1,3-dioxane-2-yl)benzonitril
5	13.7 %	4-pentylphenyl 4-methylbenzoate
	13.8 %	4-pentylphenyl 4-propylbenzoate
	7.0 %	4-pentylphenyl 4-pentylbenzoate
	4.0 %	4-propylphenyl 4-heptylbenzoate
	10.0 %	4-pentylphenyl 4-heptylbenzoate
10	12.5 %	4-heptylphenyl 4-heptylbenzoate
	11.0 %	4'-cyano-4-biphenyl 4-ethyl-1-cyclohexyl-carboxylate
	2.5 %	4'-cyano-4-biphenyl 4'-heptyl-4-biphenyl-carboxylate
15	5.0 %	4-propylphenyl 4-(4-propylbenzoyloxy)-benzoate
	9.0 %	2-fluoro-4-pentylphenyl 4'-heptyl-4-biphenyl-carboxylate

20 has a clear point of 62°C and a viscosity of 53m PaS at 20°C.

The properties of the LC compositions described above (Examples A and B) in a twisted nematic cell are compared in Table 1 with those of a commercially available LC composition ZLI-2448-000 (E. Merck, Darmstadt, Germany).

From the results given in Table I it is clearly noted that the LC compositions according to the invention (Examples A and B) have much steeper electro-optical characteristics ( $\gamma$ ).

- 5 In addition there is a clear improvement of the viewing-angle dependence of the contrast ( $\beta$ ). The achieved values of  $\gamma$  and  $\beta$  combined with the low threshold voltage  $V_{90}$  allow to realize a dot-matrix  
10 twisted nematic liquid crystal display device addressed by multiplex ratios of 1 : 100, using commercially available driver circuits.

These devices show excellent display performances and practically acceptable operating life-time.

Table I

Item	Conven- tional example	Example A of this invention	Example B of this invention
$V_{10,0,25}$ [Volt] (threshold voltage)	2,19	1,83	1,90
$\gamma = \frac{V_{50,0,25} - V_{10,0,25}}{V_{10,0,25}} \cdot 100$ [%] (steepness of the electro-optical characteristic)	15,9	12,8	13,0
$\beta = \frac{V_{10,0,25} - V_{10,40,25}}{\frac{1}{2}(V_{10,0,25} + V_{10,40,25})} \cdot 100$ [%] (viewing-angle dependence)	23,1	18,5	18,0
$k_{33}/k_{11}$ (25°C) Ratio of bend/splay elastic constants, which mainly determines the steepness of the electro-optical characteristic. The smaller the ratio $k_{33}/k_{11}$ , the steeper the electro-optical characteristic.	1,12	0,82	0.92

## Patent Claims:

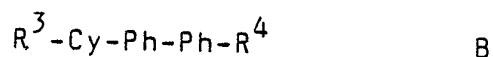
1. Liquid crystal composition, characterized in that it contains at least one compound of the formula I



wherein R is alkyl of 2 to 5 carbon atoms, Dio is trans-1,3-dioxane-2,5-diyl and Ph is 1,4-phenylene, and at least one compound of each of the following groups A to D:



wherein  $\text{R}^1$  and  $\text{R}^2$  are each alkyl of 3 to 5 carbon atoms, Cy is trans-1,4-cyclohexylene and Ph is 1,4-phenylene,

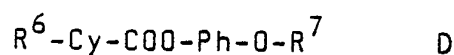


wherein  $\text{R}^3$  and  $\text{R}^4$  are each alkyl of 2 to 5 carbon atoms and Cy and Ph have the meanings indicated above,

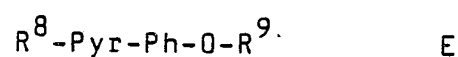
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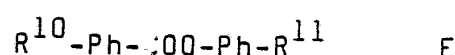
wherein  $R^5$  is alkyl of 2 to 5 carbon atoms and Cy and Ph have the meanings indicated above,



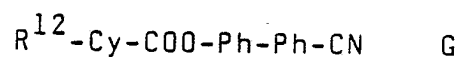
wherein  $R^6$  and  $R^7$  are each alkyl of up to 5 carbon atoms and Cy and Ph have the meanings indicated above, and, in addition in a total amount of 10 to 26% by weight at least five compounds of the formula E



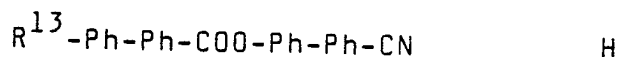
wherein  $R^8$  and  $R^9$  are each alkyl of 5 to 11 carbon atoms, Pyr is pyrimidine-2,5-diyl and Ph has the meaning indicated above, or at least one compound of each of the following groups F to K:



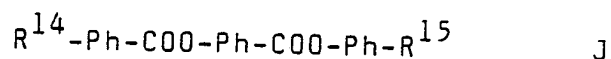
wherein  $R^{10}$  and  $R^{11}$  are each alkyl of 1 to 7 carbon atoms, Ph has the meaning indicated above,



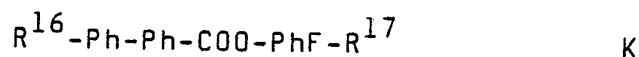
wherein  $R^{12}$  is alkyl of 1 to 3 carbon atoms, and Cy and Ph have the meaning indicated above.



wherein  $R^{13}$  is alkyl of 6 to 8 carbon atoms and Ph has the meaning indicated above.



wherein  $R^{14}$  and  $R^{15}$  are each alkyl of 1 to 5 carbon atoms and Ph has the meaning indicated above,



wherein  $R^{16}$  and  $R^{17}$  are each alkyl of 5 to 7 carbon atoms, PhF is fluorinated 1,4-phenylene and Ph has the meaning indicated above.

2. Electro-optical display device, characterized that it contains a liquid crystal composition according to Claim 1.

# INTERNATIONAL SEARCH REPORT

International Application No PCT/EP 85/00529

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>6</sup> According to International Patent Classification (IPC) or to both National Classification and IPC IPC <sup>4</sup> : C 09 K 19/42; C 09 K 19/34																				
<b>II. FIELDS SEARCHED</b> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Minimum Documentation Searched <sup>7</sup></div> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 25%; border-bottom: 1px solid black;">Classification System</td> <td style="border-bottom: 1px solid black;">Classification Symbols</td> </tr> <tr> <td style="border-bottom: 1px solid black;">IPC<sup>4</sup></td> <td style="border-bottom: 1px solid black;">C 09 K 19/</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>8</sup></div>			Classification System	Classification Symbols	IPC <sup>4</sup>	C 09 K 19/														
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<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup></b> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 10%; border-bottom: 1px solid black;">Category <sup>9</sup></th> <th style="width: 60%; border-bottom: 1px solid black;">Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup></th> <th style="width: 30%; border-bottom: 1px solid black;">Relevant to Claim No. <sup>13</sup></th> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>GB, A, 2067586 (CHISSO) 30 July 1981, see page 1, lines 49-65; example 2 --</td> <td style="text-align: center; vertical-align: top;">1,2</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>GB, A, 2080561 (HITACHI) 3 February 1982, see claims 1,6 --</td> <td style="text-align: center; vertical-align: top;">1,2</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>GB, A, 2085877 (HOFFMANN LA ROCHE) 6 May 1982, see page 14, lines 1-59; claims 1-10 --</td> <td style="text-align: center; vertical-align: top;">1,2</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>US, A, 4356104 (J.J. HSU) 26 October 1982, see example; claims 1-8 --</td> <td style="text-align: center; vertical-align: top;">1,2</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">P,Y</td> <td>EP, A, 0151446 (MERCK) 14 August 1985, see page 13, example 2  -----</td> <td style="text-align: center; vertical-align: top;">1,2</td> </tr> </table>			Category <sup>9</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>	Y	GB, A, 2067586 (CHISSO) 30 July 1981, see page 1, lines 49-65; example 2 --	1,2	Y	GB, A, 2080561 (HITACHI) 3 February 1982, see claims 1,6 --	1,2	Y	GB, A, 2085877 (HOFFMANN LA ROCHE) 6 May 1982, see page 14, lines 1-59; claims 1-10 --	1,2	Y	US, A, 4356104 (J.J. HSU) 26 October 1982, see example; claims 1-8 --	1,2	P,Y	EP, A, 0151446 (MERCK) 14 August 1985, see page 13, example 2  -----	1,2
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P,Y	EP, A, 0151446 (MERCK) 14 August 1985, see page 13, example 2  -----	1,2																		
<div style="display: flex; justify-content: space-between;"> <div style="width: 48%;"> <p><sup>10</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 48%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"Z" document member of the same patent family</p> </div> </div>																				
<b>IV. CERTIFICATION</b> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; border-bottom: 1px solid black;">Date of the Actual Completion of the International Search</td> <td style="width: 50%; border-bottom: 1px solid black;">Date of Mailing of this International Search Report</td> </tr> <tr> <td style="text-align: center;">25th February 1986</td> <td style="text-align: center;">21 MARS 1986</td> </tr> <tr> <td style="border-bottom: 1px solid black;">International Searching Authority</td> <td style="border-bottom: 1px solid black;">Signature of Authorized Officer</td> </tr> <tr> <td style="text-align: center;">EUROPEAN PATENT OFFICE</td> <td style="text-align: center;">M. VAN MOI </td> </tr> </table>			Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	25th February 1986	21 MARS 1986	International Searching Authority	Signature of Authorized Officer	EUROPEAN PATENT OFFICE	M. VAN MOI										
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# ANNEX TO THE INTERNATIONAL SEARCH REPORT ON

INTERNATIONAL APPLICATION NO.

PCT/EP 85/00529 (SA 11249)

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 10/03/86

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