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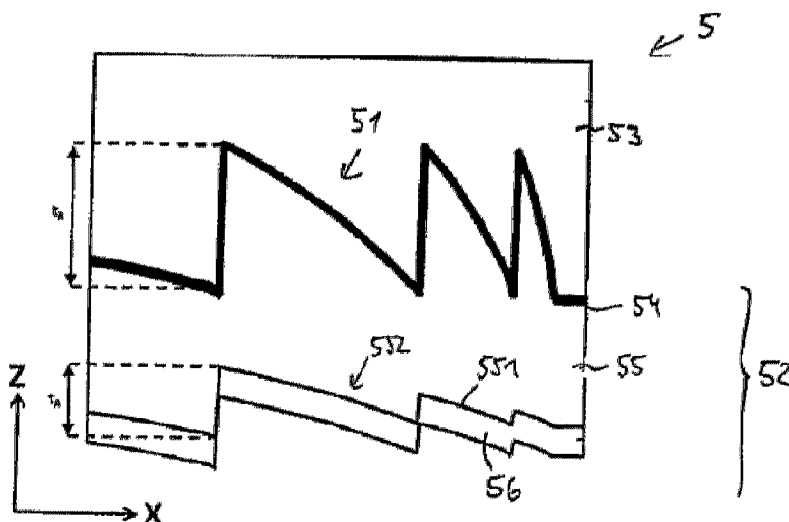
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(72) **Inventeurs/Inventors:**  
WALTER, HARALD, CH;  
BREHM, LUDWIG, DE;  
MADER, SEBASTIAN, CH;  
TOMPKIN, WAYNE ROBERT, CH;  
UNAL, ESER ALPER, DE

(73) **Propriétaires/Owners:**  
OVD KINEGRAM AG, CH; ...

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(54) **Title: MULTILAYER BODY AND METHOD FOR PRODUCING SAME**



(57) **Abrégé/Abstract:**

The invention relates to a method for producing a multilayer body, with the steps of: a) providing a substrate film with a replication layer; b) molding a surface relief appearing to the observer in the form of a three-dimensional free-form surface, which is formed in particular by structures with a lens-like design generating a magnifying, demagnifying or distorting effect, into a surface of the replication layer; c) applying a first metal layer to the surface of the replication layer forming the surface relief; d) wet-chemically applying an at least partially transparent spacer layer to the metal layer; e) applying a second metal layer to the spacer layer. The invention furthermore relates to a multilayer body produced in this way.

(73) **Propriétaires(suite)/Owners(continued):** LEONHARD KURZ STIFTUNG & CO. KG, DE

(74) **Agent:** GOWLING WLG (CANADA) LLP

## Abstract

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The invention relates to a method for producing a multilayer body, with the steps of:

- a) providing a substrate film with a replication layer;
- b) molding a surface relief appearing to the observer in the form of a three-  
10 dimensional free-form surface, which is formed in particular by structures with a lens-like design generating a magnifying, demagnifying or distorting effect, into a surface of the replication layer;
- c) applying a first metal layer to the surface of the replication layer forming the surface relief;
- 15 d) wet-chemically applying an at least partially transparent spacer layer to the metal layer;
- e) applying a second metal layer to the spacer layer.

The invention furthermore relates to a multilayer body produced in this way.  
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(Fig. 5)

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10    **Multilayer body and method for producing same**

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The invention relates to a method for producing a multilayer body, a multilayer body produced in this way, as well as a security element with such a multilayer  
15    body.

It is known to use thin-film systems in multilayer bodies which are used as security elements. For this, a semi-transparent metal layer, a transparent spacer layer and a second opaque metal layer are applied to a substrate by  
20    vacuum deposition, with the result that a so-called Fabry-Pérot system is obtained which displays a color change effect when the angle of observation or illumination is changed. The substrate can be, for example, a self-supporting single-layered or multilayer film, which can have still further applied layers, for example varnish layers.

25

Investigations have now shown that the color change effect of such a security feature displays almost the same color impression over the entire area when

observed perpendicularly or at relatively acute observation angles  $\alpha$  of from approx. 0° to 20°, even if a surface relief structure is molded into the substrate or into a varnish layer applied to the substrate.

- 5 By a surface relief structure is meant a surface relief in the form of a free-form surface which is formed in particular by structures with a lens-like design generating a magnifying, demagnifying or distorting effect. A surface relief structure is preferably formed by a surface relief in the form of a Fresnel lens structure.

10

The object of the present invention is to provide a multilayer body as well as a method for producing same, which guarantees an improved optical impression and increased security.

- 15 This object is achieved by a method, by a multilayer body as well as by a security document.

Such a method for producing a multilayer body comprises the steps of:

- a) providing a substrate film with a replication layer;
- 20 b) molding a surface relief appearing to the observer in the form of a three-dimensional free-form surface, which is formed in particular by structures with a lens-like design generating a magnifying, demagnifying or distorting effect, into a surface of the replication layer;
- c) applying a first metal layer to the surface of the replication layer forming the
- 25 surface relief;
- d) wet-chemically applying an at least partially transparent spacer layer to the

first metal layer;

e) applying a second metal layer to the spacer layer,

wherein at least one of the two metal layers is formed semi-transparent.

- 5    Optionally, one of the two metal layers can be formed as an opaque mirror layer.

A multilayer body is hereby obtained which has the following layer structure:

- a substrate film;
- a replication layer, applied to a surface of the substrate film, into the surface of
- 10    which facing away from the substrate film a surface relief appearing to the observer in the form of a three-dimensional free-form surface is introduced which comprises in particular structures with a lens-like design generating a magnifying, demagnifying or distorting effect;
- a first metal layer, which is arranged on the surface of the replication layer
- 15    forming the surface relief;
- a wet-chemically applied, at least partially transparent spacer layer, which is applied to a surface of the first metal layer facing away from the replication layer, and which optionally forms a further surface relief;
- a second metal layer, which is applied to a surface of the spacer layer facing
- 20    away from the first metal layer, wherein at least one of the two metal layers is formed semi-transparent.

Optionally, one of the two metal layers can be formed as an opaque mirror layer.

25

A layer structure is thus obtained in which a thin-film system, formed of the first metal layer, the spacer layer and the second metal layer, is combined with a surface relief. As the spacer layer is applied to the (metalized) surface relief of the replication layer, the two boundary surfaces of the spacer layer can thus  
5 each form a surface relief. These two surface reliefs are more or less correlated, as will be explained in the following.

If the transparent spacer layer is applied wet-chemically, the surface relief which forms in the surface of the transparent spacer layer facing away from the  
10 replication layer is present preferably relaxed – thus weakened. The correlation of the two surface reliefs thus decreases.

In particular the structure depth in the further surface relief is reduced compared with the surface relief of the replication layer. How strongly relaxed, i.e.  
15 weakened, the structures in the second boundary surface are depends, among other things, on the wet-chemical coating parameters, in particular the viscosity and polarity, as well as the material system, in particular polymer system, of the varnish system used, but also the solvents used. The wet and the dry layer thicknesses also have a significant influence.

20

Furthermore, measurements have shown that not only does the structure depth at the second boundary surface decrease, but the shape of the structures can sometimes also change. If, for example, a Fresnel structure with blazed grating is introduced into the replication layer, it could thus be shown that the Fresnel  
25 structures at the second boundary surface are present not only less deep, but rather also sinusoidal.

Because of the shape change of the further surface relief compared with the surface relief of the replication layer, the resulting spacer layer between these two surface reliefs thus has a variable layer thickness. As the color impression of a thin-film system is determined by the layer thickness, a multilayer body the color of which changes over its surface is thus obtained. In particular, color gradients or also color-to-gray gradients can thus be realized. Here and in the following, "gray" is to stand for uncolored or else achromatic.

Different optical effects can additionally be realized by the free-form surface of the surface relief, for example the formation of letters, numbers, but also any other objects which appear to protrude from the surface virtually or to dip behind the surface virtually. Further examples of the objects that can be represented are schematized mountains, country outlines, animals, plants or plant parts such as leaves or also buildings, bridges, gates as well as faces or portraits, in particular of famous personalities, etc. Furthermore, e.g., logos which appear as if they were bulging out and/or set back vis-à-vis the substrate surface, i.e. as if a domed surface were present in the area of the free-form surface, can be generated. The objects thus obtain spatial depth, wherein particularly characteristic optical effects which greatly increase the memorability and thus the identification effect of corresponding optically variable elements precisely in combination with the color variation generated by the thin-film system can additionally be achieved in the case of suitable formation and arrangement of partial areas with a lens-like design.

Such a multilayer body can be attached to a security document, which is preferably formed as a banknote, identification document, visa document, credit card, driver's license or the like. In this way, a visually appealing security



document that at the same time has a particularly good protection against forgery is obtained.

By an at least partially transparent spacer layer is meant here a layer with a  
5 transmittance or transmission of at least 50%, preferably at least 75% and  
particularly preferably at least 90%. The data are understood as the average of  
the transmittance or transmission over the visible wavelength range from  
430 nm to 690 nm. The wavelengths visible to humans lie in the range between  
380 nm and 780 nm of the electromagnetic spectrum, wherein the relative  
10 sensitivity of the eye in photopic vision below 430 nm and above 690 nm is  
typically less than 1% of the maximum value at 555 nm.

In a preferred embodiment the spacer layer is applied by gravure printing,  
curtain coating, slot coating, spin coating or dip coating. By means of the named  
15 methods, liquid media, in particular varnishes, can be reliably applied with a  
predetermined wet layer thickness. Some wet-chemical methods for applying  
the spacer layer are suitable for generating spacer layers with a layer thickness  
that varies in a locally targeted manner. For example, in the case of gravure  
printing, this local variation can be realized by varying the ink cell parameters on  
20 the printing roller. This makes it possible to generate two- or multi-colored  
effects in a targeted manner. Furthermore, with some wet-chemical methods  
the spacer layer can be applied not only over the whole surface, but also  
partially.

25 The spacer layer is preferably generated by applying a varnish, in particular  
based on one of the following material or polymer systems, in order thus to

achieve the desired optical properties: nitrocellulose, epoxy, polyester, rosin, acrylate, alkyd, melamine, PVA, PVC, isocyanate or urethane systems. The varnish can also consist of a mixture of two or more of the material or polymer systems.

5

Further properties of such varnishes, such as for example the viscosity or polarity, can be set to desired and advantageous values through suitable additives. Furthermore, invisible security features can also additionally be added through additives. For example, UV- or IR-active luminescent dyes, in particular fluorescent dyes, can be added to the varnishes.

10

It is furthermore expedient if a varnish with a viscosity of from 5 mPa•s to 250 mPa•s, preferably from 15 mPa•s to 200 mPa•s and particularly preferably between 20 and 170 mPa•s, is used for the application of the spacer layer.

15 With respect to the polarity of the varnish it is advantageous if the solvents of the varnish consist of solvents with a polarity index greater than 3.0 at a rate of at least 30% by mass, preferably of solvents with a polarity index greater than 3.0 at a rate of at least 50% by mass. The polarity index is understood here as in Table 2 of EP 2264491 A1.

20

The wet-chemical coating parameters, in particular the viscosity and polarity, as well as the material system, in particular polymer system, of the varnish used, but also the solvent used can influence how strongly the surface relief of the replication layer is reproduced in the surface of the spacer layer. This can influence the optical properties of the thin-film system described at the beginning.

25

The spacer layer is preferably applied with a wet layer thickness of from 1  $\mu\text{m}$  to 20  $\mu\text{m}$ , preferably from 2  $\mu\text{m}$  to 10  $\mu\text{m}$ . Both the wet and the dry layer thicknesses also influence the resulting structure depth of the relief forming in the side of the spacer layer facing away from the replication layer and thus the optical properties of the resulting thin-film system.

Furthermore it is expedient if the spacer layer is dried after application, in particular at a temperature of from 40°C to 200°C, preferably from 40°C to 150°C. There is preferably a temperature gradient in the dryer of the printing machine, wherein the temperature is between 40°C and 60°C at the start of the dryer and increases to 100°C to 150°C at the end of the dryer.

A controlled evaporation as well as a controlled curing of the varnish of the spacer layer is hereby guaranteed with, simultaneously, sufficient time for the relaxation of the surface of the spacer layer, which also has an influence on the resulting surface relief and makes a process-stable manufacture of the multilayer body with constant quality possible.

By a semi-transparent metal layer is meant a metal layer with a transmittance or transmission of at least 10%. It preferably lies between 20% and 80%, particularly preferably between 25% and 65%. Alternatively, the optical density OD is often stated instead of the transmission. The optical density is less than or equal to 1.0 in the case of the semi-transparent metal layers. It preferably lies between 0.7 and 0.1 and particularly preferably between 0.6 and 0.18. At the same time the reflectivity of the semi-transparent metal layer is further preferably at least 10% and preferably more than 15%. The data are

understood as the average of the transmittance or transmission as well as the reflectivity over the visible wavelength range from 430 nm to 690 nm.

It is furthermore preferred if the semi-transparent metal layer is generated by  
5 vacuum deposition of Cr, Al, Cu, Ti, Ni, Ag or alloys with these metals such as  
e.g. Al<sub>96%</sub>Cu<sub>4%</sub> or Inconel, in particular with a layer thickness of from 2 nm to  
20 nm, preferably from 4 nm to 15 nm.

By an opaque mirror layer is meant a metal layer with a reflectivity of at least  
10 60%, preferably more than 70% and particularly preferably more than 80%. A  
transmission of the opaque metal layer of less than 10%, preferably less than  
5%, is further preferred.

Furthermore it is preferred if the opaque mirror layer is generated by vacuum  
15 deposition of Al, Cr, Ag, Cu, or alloys with these metals such as e.g. Al<sub>96%</sub>Cu<sub>4%</sub>,  
in particular with a layer thickness of from 15 nm to 100 nm, preferably from  
20 nm to 60 nm.

The first metal layer and/or the second metal layer is expediently generated by  
20 vacuum coating or sputtering. A constant layer thickness and a good optical  
quality of the respective layer can hereby be ensured.

It is preferred if the first metal layer and/or the second metal layer is partially  
demetalized, in particular by etching and/or lift-off. In the case of structuring by  
25 etching, after application of the respective metal layer an etch resist is printed

onto the first and/or second metal layer and in the areas not covered by the etch resist the metal layer is removed by an etchant, for example a lye. The etch resist can then be removed, but it can also remain in the multilayer body and perform additional decorative functions, for example if it is a colored or  
 5 pigmented resist varnish.

The first metal layer and/or the second metal layer is preferably demetalized such that a demetalized area of the metal layer and/or of the second metal layer forms a symbol, image, logo, alphanumeric character or a combination thereof.

10

Additional items of optical information or designs can hereby be introduced into the multilayer body.

It is further preferred if the first metal layer and the second metal layer are  
 15 demetalized such that the demetalized areas overlap in the viewing direction onto the surface normal of the plane spanned by the multilayer body.

Translucent designs can hereby be implemented. A security element designed in this way allows the substrate of the security document to be seen in the overlapping demetalized areas, e.g. the printed banknote substrate or also a  
 20 window element in the substrate.

It is furthermore expedient if the first metal layer and the second metal layer are demetalized such that a metalized area of one of the metal layers overlaps the outer contour of a demetalized area of the respectively other metal layer.

25

Positioning or registration errors of the outer contour can hereby be at least partially covered by the respectively other metal layer, with the result that an appealing design that appears register-accurate results without a high outlay on positioning. By register accuracy or registration accuracy is meant a relative

5 positional accuracy between two elements, e.g. areas or layers relative to each other. This positional accuracy can be set by means of so-called register marks or registration marks, which are specifically formed such that the respective tolerance of the positional accuracy can in particular be read optically particularly well or detected by machine. These register marks or registration

10 marks can represent separate elements, which are arranged, for example, in an edge area. These register marks or registration marks can, however, also represent integral components of the multilayer body which make it possible to optically detect clearly, e.g., characteristic contours of a motif or structures of the motif or structures of the surface relief or similar.

15

It is advantageous if the surface relief and the further surface relief at the two boundary surfaces of the spacer layer at least partially correlate.

By an at least partial correlation of the reliefs is meant that the reliefs are

20 similar, but not identical, and in particular can be transferred into each other through an imaging function, for example a compression. In the simplest case the surface reliefs thus have the same structure, but different relief depths. The peaks or troughs of the relief structures further preferably lie on top of each other in each case, i.e. the peaks of the surface relief in the boundary surface

25 between the replication layer and the first metal layer, seen vertically, lie on top of the peaks of the surface relief in the boundary surface between the spacer layer and the second metal layer.

As explained at the beginning, however, structural differences are also possible, such as for example a rounding off of a blazed grating into a more sinusoidal cross-sectional structure. As already stated, the desired properties of the multilayer body are made possible precisely through this partial correlation, with  
5 the result that color progressions or color-to-gray gradients are made possible in the appearance of the multilayer body.

It is particularly advantageous if a structure depth of the further surface relief is at most 90%, preferably 80%, further preferably at most 50%, of a structure  
10 depth of the surface relief.

If the structure depth of the further surface relief is more than 50% of the structure depth of the surface relief, a color-to-gray gradient appears in the appearance of the multilayer body, otherwise a progression between two colors  
15 results.

The structure depth of the surface relief can lie between 200 nm and 20  $\mu\text{m}$ . The structure depth of the surface relief is preferably 200 nm to 2000 nm, preferably 300 nm to 1500 nm. These depth ranges are particularly suitable, in  
20 combination with the preferred values for the average dry layer thickness of the spacer layer described below, for achieving the desired optical effects.

It is furthermore preferred if the spacer layer has an average dry layer thickness of from 200 nm to 800 nm, preferably from 300 nm to 700 nm, wherein the dry  
25 layer thickness can be variable in the lateral direction.

The local dry layer thickness  $d$ , together with the refractive index  $n$  of the spacer layer as well as the observation angle  $\alpha$ , determines the resulting color impression. With a refractive index of 1.5 and an observation angle of  $30^\circ$ , for example a red color impression results if the dry layer thickness is 425 nm, a  
 5 green color impression results if the thickness is 350 nm and a blue color impression results if the thickness is 300 nm.

It is further preferred if the spacer layer has a refractive index of from 1.35 to 1.65, preferably of approximately 1.5. As already explained, the color  
 10 impression of the multilayer body can be set through the interplay of refractive index and dry layer thickness.

The surface relief advantageously has microstructures with a depth-to-width ratio of less than 0.5, preferably of less than 0.4. The microstructures can be  
 15 refractive – e.g. micromirrors – or diffractive – e.g. Fresnel-type diffraction gratings – microstructures. By width of the microstructures is meant here the distance from peak to peak or the period of the micromirrors or the diffractive microstructures.

20 The surface relief in the form of a free-form surface is preferably formed as a diffractive free-form element with a grating structure.

It is advantageous if the grating structure comprises grating lines substantially following the outlines of the free-form surface, wherein the distance between the  
 25 grating lines and/or the grating depth changes over the grating structure and in particular changes continuously from the central area of the free-form surface



out towards its edge. The period – i.e. the distance between the grating lines – and/or the depth of the surface relief varies in order to represent the desired free-form surface. Therefore it is further preferred if the depth-to-width ratio in at least 80% of the surface area of the free-form surface is less than 0.3. The

5 grating lines can, but do not have to, continuously follow the outlines of the free-form surface. They can also be interrupted, in particular interrupted with a random pattern, e.g. in order to add a scattering action to the effect of the grating structure. This can increase the observation angle for the overall effect. The grating structure can also be alternatively or additionally superimposed with

10 a structure scattering in a mat way. Both interrupted grating lines and the superimposition with a structure scattering in a mat way can lead to a more achromatic appearance of the grating structure – not of the thin-film system.

Through such a combination of a free-form surface with a grating structure, a

15 diffractive lens effect can be achieved which gives the multilayer body the desired appearance. A special feature of the diffractive free-form elements formed by grating structures is to be seen in the fact that such diffractive lens structures – unlike refractive lenses – generate a different visual impression depending on the light wavelength used for the illumination or observation of the

20 object in each case, whereby in turn particular design or security effects can be achieved.

It is preferred if a period of the grating lines at least in a partial area of the grating structure is smaller than 50  $\mu\text{m}$ , preferably smaller than 30  $\mu\text{m}$ ,

25 particularly preferably smaller than 15  $\mu\text{m}$ .

- The local dry layer thickness can be influenced in particular by a variation of the period. In the case of a varnish system which leads to a decoupling of the boundary surfaces – thus of the two surface reliefs – a further effect can also occur in addition to the shape change or shape leveling. Presumably, during the drying process of the wet-chemically applied spacer layer, capillary forces which originate from the grating grooves of the surface relief push material of the spacer layer in the direction of a larger period of the structures – typically in the direction of the center of the surface relief. The dry layer thickness is thereby greater in the center, or in areas with a coarse period  $\Delta x$ , than at the edge, or in areas with a smaller period  $\Delta x$  of the relief structures. The resulting color gradient is continuous and perfectly registered relative to the shape of the surface relief. The area with the second color typically starts when the period of the microstructures is  $\leq 30 \mu\text{m}$ , preferably  $\leq 20 \mu\text{m}$ , in particular  $\leq 15 \mu\text{m}$ .
- 15 It is further preferred if the substrate film consists of a flexible plastic film. For example, the substrate film consists of PET, PEN, MOPP, PP, PA, PC, COC or PVC.

- 20 The substrate film expediently has a layer thickness of from  $4 \mu\text{m}$  to  $500 \mu\text{m}$ , preferably from  $8 \mu\text{m}$  to  $150 \mu\text{m}$  and particularly preferably from  $12 \mu\text{m}$  to  $50 \mu\text{m}$ .

- 25 It is further advantageous if the multilayer body has at least one further translucent varnish layer, which is dyed by means of at least one dye and/or pigment. The at least one dye is preferably a soluble dye or luminescent substance. Dyes from the following substance groups are preferably used:

metal complex dyes, in particular with  $\text{Cr}^{3+}$  or  $\text{Co}^{2+}$  as the central atom.

Examples are the Neozapon dyes from BASF and Orasol dyes from BASF (formerly CIBA).

- 5 Further color effects can hereby be achieved, or the color effects of the thin-film system can be filtered and modulated. For example, a red color impression of the thin-film system can be modulated to an orange color impression through a translucent varnish layer dyed yellow.
- 10 It is expedient in particular if the at least one further varnish layer is arranged between the replication layer and the substrate film.

Additionally or alternatively, the replication layer can also be dyed by means of a dye and/or pigment, in order also to generate the named color effects.

15

- Additionally or alternatively, the surface relief in the form of a free-form surface can also be combined with structures generating other effects. For example, the surface relief can also be gridded with a 2D, or 2D/3D or 3D hologram, in particular combined in an interlaced arrangement, which represents a number
- 20 floating over the free-form surface. The proportion of surface area of the structures generating other effects is preferably small, in order to influence the color effect of the surface relief in the form of a free-form surface in combination with the thin-film setup as little as possible. The proportion of surface area is preferably less than 30%, particularly preferably less than 20% and in particular
- 25 preferably  $\leq 10\%$ .

The multilayer body can be implemented as a transfer film or as a laminating film. In the case of a transfer film the substrate film is joined to the remaining layers of the multilayer body, which form a transfer ply, by means of a detachment layer. In the case of the application of the multilayer body to a substrate, the multilayer body is secured, in particular glued, to the substrate and then the substrate film is peeled off the applied transfer ply. The application to the substrate can be effected, for example, by means of hot embossing or by means of cold embossing. In the case of a laminating film, the substrate film also remains on the multilayer body after the application of the multilayer body to a substrate.

The invention and embodiments thereof are explained in more detail below with reference to the drawing. There are shown in:

- 15    Fig. 1            a schematic illustration of the mode of operation of a Fabry-Pérot thin-film system according to the state of the art;
  
- Fig. 2            a schematic representation of a three-dimensional free-form surface in top view and in cross section;
  
- 20    Fig. 3            a schematic detail view of a surface relief which optically imitates the three-dimensional free-form surface shown in Fig. 2 as well as an enlarged section of the surface relief;
  
- Fig. 4            a schematic sectional representation through a multilayer body with a surface relief and a thin-film system according to the state of the art;
- 25

- Fig. 5      a schematic sectional representation through an embodiment example of a multilayer body with a surface relief and a thin-film system with a wet-chemically deposited spacer layer;
- 5      Fig. 6      a schematic sectional representation through a further embodiment example of a multilayer body with a surface relief and a thin-film system with a wet-chemically deposited spacer layer;
- 10      Fig. 7      a schematic sectional representation through a further embodiment example of a multilayer body with a surface relief and a thin-film system with a wet-chemically deposited spacer layer;
- 15      Fig. 8      a schematic sectional representation through a further embodiment example of a multilayer body with a surface relief and a thin-film system with a wet-chemically deposited spacer layer;
- Fig. 9      reflection spectra of multilayer bodies with a surface relief and a thin-film system;
- 20      Fig. 10      a schematic sectional representation through a further embodiment example of a multilayer body with a surface relief, a color layer and a thin-film system with a wet-chemically deposited spacer layer;

- Fig. 11 a schematic sectional representation through a further embodiment example of a multilayer body with a surface relief, a partial metal layer and a thin-film system with a wet-chemically deposited spacer layer;
- 5
- Fig. 12 a schematic view of a security document with a multilayer body according to Fig. 11;
- 10
- Fig. 13 a schematic view of a motif realized by means of a multilayer body according to Fig. 11;
- Fig. 14 a schematic representation of the method steps for producing an embodiment example of a multilayer body with partial demetalization;
- 15
- Fig. 15 a security element produced by means of a method according to Fig. 14 in front view;
- 20
- Fig. 16 a schematic representation of the method steps for producing an alternative embodiment example of a multilayer body with partial demetalization;
- Fig. 17 a security element produced by means of a method according to Fig. 16 in front view;

Fig. 18 a schematic representation of the method steps for producing a further alternative embodiment example of a multilayer body with partial demetalization;

5 Fig. 19 a schematic representation of the method steps for producing a further alternative embodiment example of a multilayer body with partial demetalization.

A Fabry-Pérot thin-film system 1 represented schematically in Fig. 1 consists of  
 10 a semi-transparent layer 11, usually a first metal layer (e.g. Cr, Al, Cu, Ti, Ag, Ni or Inconel), a transparent spacer layer 12 (e.g.  $\text{MgF}_2$ ,  $\text{SiO}_2$  or a polymer varnish) as well as a second metal layer 13. The latter likewise usually consists of metal (e.g. Al, Cr, Ag, Cu). It can be opaque or also semi-transparent. The layers 11, 12, 13 are deposited onto a substrate film 2. The sequence of the  
 15 layers 11, 12 and 13 can also be reversed and the effect can be visible from the back, e.g. through a window of a security document.

Part of the light incident at the angle  $\alpha_{\text{in}}$  is reflected on the semi-transparent first metal layer 11, part is absorbed and part is transmitted. The transmitted part  
 20 penetrates the transparent spacer layer 12 and is reflected on the second metal layer 13. The reflected part of the light again penetrates the transparent spacer layer 12 and is partially transmitted through the semi-transparent layer 11. This transmitted part interferes with the portion of the light reflected on the semi-transparent layer 11 and reaches the eye of the observer at the angle  $\alpha_{\text{out}}$ ,  
 25 wherein  $\alpha_{\text{in}} = \alpha_{\text{out}} = \alpha$ . Light refraction was disregarded in this schematic representation. Because of the interference of the light portions, the light appears colored to the observer. The color impression here depends among

other things on the thickness  $d$  as well as on the refractive index  $n$  of the spacer layer 12 and the angle  $\alpha$ . Constructive interference occurs when the optical path difference  $\Delta$  between the portion of the light reflected on the first metal layer 11 and the portion of the light reflected on the second metal layer 13 is a multiple  
 5 of the wavelength  $\lambda$ .

There are various coating methods which can be used to produce such a layer system 1. The two metal layers 11, 13 are usually applied using vacuum technology, e.g. by means of evaporation or sputtering. In contrast, the  
 10 transparent spacer layer 12 can on the one hand be applied using vacuum technology, e.g. by means of thermal evaporation of  $\text{MgF}_2$ . On the other hand, such transparent layers 12 can also be produced wet-chemically, e.g. by means of gravure printing or slot coating.

15 These different production methods lead to very similar results on smooth substrates 2; a surface with a homogeneous color tilt effect or color change effect is achieved. On structured surfaces, in contrast, wet-chemically applied transparent spacer layers 12 result in clearly different layer patterns from spacer layers 12 applied using vacuum technology. This is due to the fact, among other  
 20 things, that layers applied using vacuum technology are built up piece by piece, while wet-chemically applied layers are usually applied all at once and the liquid layer only cures in the drying and/or curing process, e.g. UV curing.

Consequently, such thin-film systems 1 can be applied not only to the smooth  
 25 surfaces shown in Fig. 1, but also to structured surfaces. An example of this is shown in Fig. 2. Surface relief structures 3 generate a three-dimensional surface which seems to protrude from the substrate plane. The shape of this 3D surface can be chosen almost randomly (free-form). In Fig. 2 the surface relief



structure 3 imitates a droplet with an optically three-dimensional effect as a free-form surface. The structures are a special variant of Fresnel lenses. Fig. 2a shows a schematic top view of such a surface relief structure 3, which imitates the 3D droplet. An exemplary section through this three-dimensional shape is indicated with A-A'.

Fig. 2b shows a schematic side view along the section A-A' through this three-dimensional shape, wherein here the height profile 31 seemingly generated (not really present) is represented in the  $z'$  direction.

10

Such a three-dimensional surface shape or an apparent height profile 31 can be optically generated by Fresnel lens-type microstructures 32. Fig. 3a shows a schematic side view (not to scale) of exemplary Fresnel-type relief structures 32 which optically generate the three-dimensional droplet shape from Fig. 2. For simplification, only a few grooves of the Fresnel lens are represented. If the representation were to scale, many more would have to be represented. Furthermore, to simplify the representation, the  $z$ -scale is reproduced greatly extended. In addition to the continuous Fresnel structures represented, so-called binary Fresnel structures can also generate a similar effect. In other words, the comparatively strongly 3D domed impression from Figs. 2a, 2b can be simulated with special diffractive structures the extent of which in the  $z$  direction can be significantly smaller than simulated by the achieved optical effect.

25 Fig. 3b shows the area marked by a dotted circle in Fig. 3a enlarged and represented approximately to scale. In the currently used hot-embossing films, the smallest period or structure width  $\Delta x_{\min}^{\text{I}}$  of the surface relief structures 32 is approx. 3  $\mu\text{m}$  and thus is approximately two to three times larger than the depth

$t_R$ . The average depth-to-width ratio of the structure elements is consequently smaller than 0.5. Here the depth of the structures  $t_R$  typically lies in the range from 500 nm to 1500 nm. The angles of the edges 321 of the relief structures 32 in reality are consequently much less acute than Fig. 3a shows.

5

A schematic cross section, not to scale, through a multilayer body 4 known from the state of the art is shown in Fig. 4. This comprises a substrate film 41 with a replication layer 42, into which a surface relief 43 is introduced. To this, in each case using vacuum technology, a semi-transparent first metal layer 44, a  
 10 transparent spacer layer 45 and a second metal layer 46 are applied, which form a thin-film system 47 of the described type. The replicated surface relief 43 reproduces itself recognizably in the further layers. It is to be recognized that the replicated structures are present almost identically in the second boundary surface of the transparent spacer layer 45 – thus the boundary surface between  
 15 the spacer layer 45 and the second metal layer 46. This is the case with spacer layers 45 applied purely using vacuum technology, as here these layers are built up piece by piece. The structure depth  $t_A$  on the side of the spacer layer 45 opposite the replication varnish usually lies – in the case of the typical layer thicknesses of the spacer layer 45 applied using vacuum technology – in the  
 20 range from 90% to 100% of the original depth in the replication varnish  $t_R$ . The peaks of the structures here can, however, also be present slightly rounded.

When observed perpendicularly or at relatively acute observation angles  $\alpha$  of from approx.  $0^\circ$  to  $20^\circ$ , the color effect of such a security feature - consisting of  
 25 the relief structures 43 of the replication layer 42, coated with the thin-film system 47 - displays almost the same color impression over the entire area of the relief structures, as the layer thickness overall is almost identical. The thin-film filter thus acts as in a flat – i.e. unstructured – area.

Fig. 5 shows, not to scale, an embodiment example of a multilayer body 5 which combines a surface relief 51 with a thin-film system 52. The multilayer body 5 comprises a substrate film 53, which preferably consists of PET, PEN, MOPP, PP, PA, PC, COC or PVC and has a layer thickness of from 4  $\mu\text{m}$  to 500  $\mu\text{m}$ ,  
5 preferably from 8  $\mu\text{m}$  to 150  $\mu\text{m}$  and particularly preferably from 12  $\mu\text{m}$  to 50  $\mu\text{m}$ .

A surface relief 51 is introduced into the substrate film 53 or into a replication layer applied to the substrate film 53. This is preferably formed as a diffractive  
10 free-form element with a grating structure.

It is advantageous if the grating structure comprises grating lines substantially following the outlines of the free-form surface, wherein the distance between the grating lines and/or the depths of the grating structure changes over the grating  
15 structure and in particular changes continuously from the central area of the free-form surface out towards its edge.

Through such a combination of a free-form surface with a grating structure, a diffractive lens effect can be achieved which gives the multilayer body the  
20 desired appearance.

It is preferred if a period of the grating lines at least in a partial area of the grating structure is smaller than 50  $\mu\text{m}$ , preferably smaller than 30  $\mu\text{m}$ , particularly preferably smaller than 15  $\mu\text{m}$ .

25

A semi-transparent metal layer 54 is applied to the surface relief 51, preferably by vacuum coating, sputtering or the like. It is preferred if the metal layer consists of Cr, Al, Cu, Ti, Ni, Ag or Inconel and has a layer thickness of from 2 nm to 20 nm, preferably from 4 nm to 15 nm.

5

A transparent spacer layer 55 is applied to the side of the metal layer 54 facing away from the surface relief 51 wet-chemically, in particular by means of gravure printing, curtain coating, slot coating or the like.

- 10 The spacer layer 55 is preferably generated by application of a varnish, in particular based on nitrocellulose, epoxy, polyester, rosin, acrylate, alkyd, melamine, PVA, PVC, isocyanate or urethane systems, in order thus to generate the desired optical properties.
- 15 The varnish can contain further additives, such as e.g. crosslinkers or surfactants, in order to set its physico-chemical properties, such as for example the viscosity or polarity, to desired and advantageous values.

- 20 It is furthermore expedient if, for application of the spacer layer, a varnish with a viscosity of from 5 mPa•s to 250 mPa•s, preferably from 15 mPa•s to 200 mPa•s and particularly preferably between 20 mPa•s and 170 mPa•s is used. With respect to the polarity of the varnish, it is advantageous if the solvents of the varnish consist of solvent with a polarity index greater than 3.0 at a rate of at least 30% by mass, preferably of solvent with a polarity index  
25 greater than 3.0 at a rate of at least 50% by mass. The polarity index here is understood as in Table 2 of EP 2264491 A1.

The surface relief 51 of the substrate film 53 reproduces itself on the side of the spacer layer 55 facing away from the surface relief 51. The wet-chemical coating parameters, in particular the viscosity and polarity of the varnish system used, but also the varnish system used and/or the solvent or solvents, can  
5 influence how strongly the surface relief 51 reproduces itself in the surface 551 of the spacer layer 55.

The spacer layer 55 is preferably applied with a wet layer thickness of from 1  $\mu\text{m}$  to 20  $\mu\text{m}$ , preferably from 2  $\mu\text{m}$  to 10  $\mu\text{m}$ . Both the wet and the dry layer  
10 thicknesses also influence the resulting structure depth of the relief 552 forming in the side of the spacer layer 55 facing away from the replication layer 53 and thus the optical properties of the resulting thin-film system 52.

After the wet-chemical deposition, the spacer layer 55 is dried, in particular at a  
15 temperature of from 40°C to 200°C, preferably from 40°C to 150°C. There is preferably a temperature gradient in the dryer of the printing machine, wherein the temperature is between 40°C and 60°C at the start of the dryer and increases to 100°C to 150°C at the end of the dryer. This results in a controlled average dry layer thickness of from 200 nm to 800 nm, preferably from 300 nm  
20 to 700 nm, with the desired degree of correlation between the two surface reliefs 51, 552.

Finally, a second metal layer 56, preferably made of Al, Cr, Ag or Cu with a preferred layer thickness of from 15 nm to 100 nm, particularly preferably from  
25 20 nm to 60 nm, is also deposited onto the side of the spacer layer 55 facing away from the metal layer 54.

- Because of the wet-chemical deposition, the structure depth  $t_A$  of the surface relief 552 reproduced in the spacer layer 55 decreases appreciably compared with the structure depth  $t_R$  of the surface relief 52 in the substrate film 53, i.e.  $t_A \leq 80\% t_R$ . How strongly relaxed, i.e. weakened, the structures in the second
- 5 boundary surface are depends, among other things, on the wet-chemical coating parameters, in particular the material base, the viscosity and the polarity of the varnish system used, but also the solvents used. The wet and the dry layer thicknesses also have a significant influence.
- 10 Measurements have shown that not only does the structure depth in the relief 552 of the spacer layer 55 decrease, but the shape of the structures can sometimes also change. Micrographs thus show that, for example, a blazed shape of Fresnel structures present at the first boundary surface – thus between the semi-transparent metal layer 54 and the spacer layer 55 – is
- 15 present not only less deep, but rather also sinusoidal at the second boundary surface – thus between the spacer layer 55 and the second metal layer 56. This is represented schematically in Fig. 6.

- Because of this shape leveling and/or the decreasing correlation between the
- 20 two surface reliefs 51, 552, the thickness of the spacer layer can vary relatively greatly, which then leads to a substantial weakening of the color contrast. As the color impression is influenced directly by the layer thickness of the spacer layer 55, such a relatively greatly varying layer thickness results in a greatly varying color impression in a lateral extent in the size of the grating period and
- 25 thus below the resolution limit of the human eye.

For example, at an observation angle of  $30^\circ$ , a layer thickness of the printed spacer layer 55 (refractive index  $n=1.5$ ) of 425 nm yields a red color impression,

a thickness of 350 nm yields a green color impression and a thickness of 300 nm yields a blue color impression. The mixed color of such a spacer layer 55 with a layer thickness varying greatly below the resolution limit of the human eye therefore usually appears dark gray or metallic gray to the human eye. The

5 greatly varying layer thickness of the spacer layer 55 is mainly present with a fine grating period in areas of the relief structure 51, 552. In this case there is a gradient from color in areas with a coarse period of the relief structure towards gray/achromatic in areas with a fine period of the relief structure. As the gradient is generated by the structures of the surface relief 51, the color-to-gray

10 gradient is perfectly registered relative to the 3D impression or the three-dimensional surface shape of the relief structures 51, 552.

The great variation of the layer thickness of the spacer layer 55 is mainly present in regions in which the period of the surface relief microstructures is

15 small – i.e.  $\leq 50 \mu\text{m}$ , preferably  $\leq 30 \mu\text{m}$ , in particular  $\leq 20 \mu\text{m}$ . The resulting variation of the layer thickness of the spacer layer 55 results in a color-to-gray gradient, wherein the area appearing gray typically starts when the period of the surface relief microstructures is  $\leq 50 \mu\text{m}$ , preferably  $\leq 30 \mu\text{m}$ , in particular  $\leq 20 \mu\text{m}$ . The color-to-gray gradient optically clearly emphasizes the free-form

20 surface.

The period of the microstructures is typically smaller towards the edge of the free-form surface, which is why the color-to-gray gradient usually runs from the center of the free-form surface to the edge.

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In addition to a color progression or color gradient from a first color to a gray or metallic impression, it is also possible to realize a progression from a first to a second color. For this it is preferred for the correlation of the two boundary

surfaces of the spacer layer 55 to be very small, i.e.  $t_A \leq 50\% t_R$  and in particular  $t_A \leq 30\% t_R$ . In the extreme case, the second boundary surface is even formed almost smooth towards the second metal layer, or the correlation between the surface reliefs 51, 552 is (almost) completely removed. This is an extreme form of the shape change or shape leveling. Figures 7 and 8 show this in schematic side views that are not to scale.

In particular in the case of a varnish system which leads to a decoupling of the boundary surfaces, a further effect can also occur in addition to the shape change or shape leveling. Presumably, during the drying process of the wet-chemically applied spacer layer 55, capillary forces which originate from the relief grooves push material of the spacer layer 55 in the direction of a larger period of the structures of the surface relief – i.e. typically in the direction of the center of the relief shape or the desired free-form surface. Furthermore, material of the spacer layer 55 is no doubt also pushed in the direction of mirror surfaces, surrounding the surface relief, or also surrounding surfaces with flatter other structures. Diffraction gratings with periods below  $2 \mu\text{m}$  and grating depths below  $200 \text{ nm}$  are an example of these other structures. The layer thickness is thereby larger in the center of the free-form surface, or in areas with a coarse period  $\Delta x$ , as well as in the areas surrounding the surface relief than at the edge of the free-form surface, or in areas with a smaller period  $\Delta x$  of the relief structures. In areas with a relatively large local period  $\Delta x$  of the relief structures the layer thickness is almost identical to the thickness in areas without structures, i.e. on a smooth surface, or almost identical to the thickness in areas with flatter other structures. The color gradient is continuous and perfectly registered relative to the shape of the surface relief. The color gradient thereby optically clearly emphasizes the free-form surface. The area with the second



color typically starts when the period of the surface relief microstructures is  $\leq 30 \mu\text{m}$ , preferably  $\leq 20 \mu\text{m}$ , in particular  $\leq 15 \mu\text{m}$ .

Multilayer bodies in the manner of the invention, which have a gradient from a first color to a second color, could even have two different color effects in the area with the surface relief in the form of a free-form surface. Thus, for example in the area with a coarse period  $\Delta x$ , there can be a color effect from red in the case of almost perpendicular observation to green in the case of tilted observation (e.g.  $30^\circ$ ). At the same time, in the area with a fine period  $\Delta x$ , there can be a color effect from green in the case of almost perpendicular observation to blue in the case of tilted observation (e.g.  $30^\circ$ ).

As a whole, it is thus possible to realize both a color-to-gray and a color-to-color progression.

A color progression from a first color to a metallic/gray/achromatic impression occurs when the relaxation is less than 50%, i.e. when  $t_A \geq 50\% t_R$ . At the same time, however,  $t_A$  must be  $\leq 90\% t_R$  and preferably  $t_A \leq 80\% t_R$ . In other words, if the structure depth at the boundary surface between the spacer layer and the second metal layer lies between 50% and 90%, preferably 50% to 80%, of the structure depth at the boundary surface between the spacer layer and the first metal layer, a color/gray gradient results.

A color progression from a first to a second color occurs above all when the structure depth relaxes by more than 50%, i.e. when  $t_A \leq 50\% t_R$ .  $t_A \leq 40\% t_R$  is preferred,  $t_A \leq 30\% t_R$  is further preferred and  $t_A \leq 20\% t_R$  is preferred in particular. In other words, if the structure depth at the boundary surface between the spacer layer and the second metal layer is smaller than/equal to

50%, preferably smaller than/equal to 40%, particularly preferably smaller than/equal to 30% and in particular smaller than/equal to 20% of the structure depth at the boundary surface between the spacer layer and the first metal layer, a gradient between two colors results.

5

The described cases can be distinguished by means of spectrometer measurements. Figs. 9A to 9C show examples of measured spectra in the case of perpendicular incidence – i.e. when  $\alpha=0^\circ$  – for the case shown in Fig. 4 of very strong correlation of the boundary surfaces in the case of a spacer layer applied using vacuum technology according to the state of the art (Fig. 9A), the case shown in Fig. 5 of weakened correlation of the surface reliefs in the case of wet-chemical deposition of the spacer layer (Fig. 9B) and the case shown in Fig. 7 of very low correlation of the surface reliefs in the case of likewise wet-chemical deposition of the spacer layer (Fig. 9C). One spectrum in the area with a coarse period of the surface relief (center, dashed-line graph) and one in the area with color gradient or in the area with a small period of the surface reliefs (edge, continuous-line graph) are shown in each case.

As shown in Fig. 9A, the spectra in the central and edge areas of the surface relief in the case of a vapor-deposited spacer layer substantially follow the same progression – i.e. the peak maxima and minima lie at the same wavelength. The thin-film system therefore appears monochromatic over the entire area of the free-form surface. The slightly reduced reflection in the edge area is due to the fact, among other things, that the surface relief diffracts part of the light more strongly out of the viewing direction of the spectrometer in the edge area than in the central area.

In Fig. 9B it is to be recognized that in the area of the surface relief 51 there is a gradient from a first color (center, dashed-line graph) to a gray/metallic impression (edge, continuous-line graph), wherein the difference  $\Delta R$  between the maximum  $R_{\max}$  and the minimum  $R_{\min}$  of the reflection measured in the case of perpendicular incidence in the spectral range from 430 nm to 690 nm is at least 3x, preferably at least 5x and in particular preferably at least 7x, greater in the area of the first color than in the area with the gray/metallic impression.

In the case of Fig. 9C it is to be recognized that in the area of the surface relief 51 there is a gradient from a first (center, dashed-line graph) to a second color (edge, continuous-line graph), wherein the two reflection spectra, belonging to the colors and measured in the case of perpendicular incidence, exhibit a shift of the wavelength  $\Delta\lambda$  of at least one of the reflection peaks of at least 10 nm, preferably at least 20 nm and in particular at least 30 nm. This reflection peak here usually lies in the spectral range from 430 nm to 690 nm. In the measured example shown in Fig. 9c, the shift of the peak wavelength is  $\Delta\lambda = 531 \text{ nm} - 508 \text{ nm} = 23 \text{ nm}$ .

Instead of the Fabry-Pérot systems described up to now, other thin-film systems can also be used, e.g. HLH systems (HLH, LHL, HLHL, LHLH, etc.). A specific example of this is a vapor deposition of the replicated surface structure with a layer of high-refractive-index material (H, H for high refractive index), e.g. 80 nm  $\text{TiO}_2$  with  $n \approx 2.54$  at  $\lambda = 589 \text{ nm}$ ,  $\text{ZrO}_2$  or  $\text{ZnS}$ . A rather low-refractive-index spacer layer (L, L for low refractive index) with the thickness  $d$  is then applied wet-chemically, e.g. 500 nm PVC varnish with  $n = 1.545$  at  $\lambda = 589 \text{ nm}$ . This layer generates the optical path difference for the interference of the light portions. In this spacer layer the structures in the second boundary surface are again present relaxed, which leads to a gradient. As a third layer, again a layer

- of high-refractive-index material (H), e.g. 80 nm  $\text{TiO}_2$ ,  $\text{ZrO}_2$  or  $\text{ZnS}$ , is applied, e.g. vapor-deposited. The thickness of the high-refractive-index layers is preferably in the range from 10 nm to 200 nm and in particular between 20 nm and 100 nm. Such a thin-film system also generates, through interplay with the relief structures, a color impression with a color progression or color gradient. The advantage of such a thin-film system is that it is transparent, and thus can also be placed over vital, in particular personalized and/or individualized, data for example in ID documents.
- 10 The color progression of the multilayer bodies 5 described up to now can also be modified by means of color filters which, seen starting from the observer, are located above the thin-film stack. For example, in the layer stack, one or more layers can be dyed.
- 15 Fig. 10 shows, in a schematic side view, the example of a dyed multilayer body 5. The layer structure corresponds to that shown in Fig. 5. An additional dyed varnish layer 58 with a preferred layer thickness of from 200 nm to 5  $\mu\text{m}$  is provided between the replication layer 53 and the substrate film 57. Alternatively, the surface relief structure 51 can, however, also be replicated
- 20 into a dyed replication varnish 53. The color impression of the surface relief 51 in all of these cases is superimposed with the color impression of the color filter 58.
- A specific example is a so-called gold foil, which has a replication varnish 53 indirectly or directly dyed yellow. In such a film, the background which is deep red for example at an observation angle of  $30^\circ$  as well as the deep red area of the surface relief would appear more orange. The chrome-metallic edge area of the surface relief on the other hand would have a dark yellow effect. The color

gradient would therefore no longer be deep red to chrome-metallic, but orange to yellow.

5 A specific example of the integration of the combination of surface relief 51 in the form of a free-form surface and thin-film system 52 in the manner of the invention into a security element 6 with a KINEGRAM® is represented in Fig. 11.

10 Here a KINEGRAM design is replicated and vapor-deposited over the whole surface with a first aluminum layer 61. Then, in a standard demetalizing process, in register, the aluminum is removed in the area 62 with the surface relief 51 as well as in the areas 63 where the thin-film effect is otherwise still to be seen. The thin-film system 52, which corresponds to the already described embodiment examples, is then applied to the back, seen from the observer, of the first aluminum layer 61.

15 Fig. 12 shows an exemplary model banknote 7 with such a demetalization design. A security strip 71 contains, among other things, a surface relief 51, which represents a free-form surface in the form of a lion and which has one of the described color gradients. This surface relief 51 is present in a demetalized area 62 of the strip. The rest of the strip has yet other effects, e.g. diffractive effects. Furthermore, the strip contains three demetalized areas 63 in the form of the number "55", wherein the Fabry-Pérot three-layer system 52 is also present in these demetalized areas. The three numbers "55" consequently display the same or a very similar color tilt effect as is also present in the surface relief 51 in areas with a coarse period. Alternatively, the color effect in the areas 63 can be different from that in the area 62. For this, a different thickness of the spacer layer 55 must be present locally. In printing processes it is easily possible to generate locally different thicknesses in a targeted manner.

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This can be achieved, e.g., by different depths of the ink cells in a gravure printing roller in the areas of the roller which deposit the spacer layer onto the areas 62 and 63.

- 5 A further alternative provides that the metal layers of the thin-film system 52 are additionally removed in the areas 63 by means of one or more further demetalizing steps. This can be achieved, e.g., via a so-called lift-off process. In this case, a color effect is present in the area 62, whereas the substrate is to be seen undisturbed by metal layers in the areas 63.

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Fig. 13 shows a schematic top view of such a security element 71 in detail. The color progression in the area of the surface relief 51 is illustrated by contour lines. The background for the motif formed by the surface relief 51 is formed by metalized surfaces 72.

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The color effect of the surface relief 51 in the form of a free-form surface can, as previously described, be visible from the front of the substrate, i.e. from the side to which the multilayer body is applied. Alternatively, the layer structure can also be designed such that the color effect is to be seen through a window in the security document to which the multilayer body is applied. In other words, an observer sees the color effect of the surface relief when he observes the back of the security document. At the same time, the multilayer body can be designed such that the observer sees the surface relief 51 in the form of a free-form surface in a metallically reflecting and spatially mirrored form on the front of the security document. The mirroring here can be not only a left/right mirroring. The free-form surface can additionally also appear bulging out on one side of the security document and optically appear to be set back on the other

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side. This increases the memorability and thus the identification effect additionally.

Fig. 14 shows an alternative method for producing a multilayer body 5 with partial demetalization of the metal layers 54, 56. A first metal layer 54, as already explained, is here also applied to the substrate film 53 with a surface relief 51 of the described type. In this embodiment example, the first metal layer 54 is formed as an opaque mirror layer. This is overprinted with an etch resist 8, which remains omitted in areas 541 to be demetalized. In these areas 541, the metal layer 54 can then be removed by etching (Fig. 14A).

After the etching, the etch resist 8 can be removed again by suitable solvents (Fig. 14B) and the uncovered metal layer 54 can be overprinted with the transparent spacer layer (Fig. 14C).

Before the deposition of the second metal layer 56, a lift-off varnish 9 is deposited onto the spacer layer 55 in those areas 561 in which the second metal layer 56 is to be removed later (Fig. 14D). The second metal layer 56 is then applied over the whole surface (Fig. 14E). In this example the second metal layer is formed semi-transparent. When the lift-off varnish 9 is removed by a suitable solvent, the second metal layer 56 is also removed with it in the areas 561 (Fig. 14F).

Demetalized areas 541 and 561 are thus obtained in the two metal layers, which overlap and which can form a motif. There is no thin-film effect in the demetalized areas 541 and 561.

If the multilayer body 5 is applied to a security document, for example a banknote 7 by means of a suitable adhesive layer 57 (Fig. 14G), the

demetalized areas allow the substrate of the banknote to be viewed. The demetalized area 541 forms, as is to be recognized in Fig. 15, the number "55", while the demetalized area 561 frames the surface relief.

- 5 The area of the surface relief 51 is arranged over a window 72 of the banknote, with the result that the effect of the surface relief 51 with the color change effect generated by the thin-film system 52 is to be recognized from the other side of the banknote 7. The demetalized areas 541 and 561, in contrast, are concealed by the substrate of the banknote 7 in the case of observation through the  
10 window 72.

- Fig. 16 shows a further alternative method for producing a multilayer body 5 with partial demetalization of the metal layers 54, 56. A first metal layer 54, as already explained, is here also applied to the substrate film 53 with a surface  
15 relief 51 of the described type. This is overprinted with an etch resist 8, which remains omitted in areas 541 to be demetalized. In these areas 541 the metal layer 54 can then be removed by etching (Fig. 16A). In addition the area of the surface relief 51 also initially remains left open here. In this embodiment the etch resist 8 is not removed and can take on additional decorative functions.

- 20 Now a lift-off varnish 9 is printed on in the areas 541 to be left open (Fig. 16B). Then a further, semi-transparent metal layer 58, preferably made of chromium, is vapor-deposited over the whole surface (Fig. 16C) and removed again by peeling off the lift-off varnish 9 in the areas 541 (Fig. 16D). The area of the  
25 surface relief 51 is thus now also metalized.

Analogously to the already described methods, the spacer layer 55 is now applied (Fig. 16E) and vapor-deposited over the whole surface with metal (Fig.



16F). An etch resist 8 is again partially printed onto the thus-generated second metal layer 56, which is opaque in this embodiment example, (Fig. 16G) and the metal layer 56 is removed in the left-open areas 561 by etching (Fig. 16H). Here too, the areas 541 and 561 again overlap.

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The multilayer body can now be applied to a banknote 7 again by means of an adhesive layer 57 (Fig. 16I). In this embodiment the substrate film 53 is removed (hot-embossing variant) and no window is provided. The multilayer body 5 displays the desired effect in the case of direct observation from the side  
 10 of the substrate film 53. As is to be recognized in Fig. 17, the demetalized area 541 again forms the number "55". The demetalized area 561 forms a frame for the surface relief 51, which is, however, covered by the metal layer 54. The oval shape of the multilayer body can be generated by applying the multilayer body 5 to the banknote 7 by means of hot-embossing with a hot-embossing die  
 15 correspondingly shaped to the oval outer contour. The remaining layers of the multilayer body 5 are peeled off the substrate film 53, and together applied to the banknote 7 as a transfer ply, only in the area where the oval hot-embossing die acts on the substrate film 53 by means of pressure and heat and activates the adhesive layer 57 of the multilayer body 5 preferably formed as a heat-  
 20 activatable adhesive layer.

A further method for producing a multilayer body 5 with partial demetalization of the metal layers is represented in schematic – not to scale – side views in Figs. 18A to 18G.

25 An opaque metal layer 58 is also applied here to the substrate film 53 with a surface relief 51 of the described type. This is overprinted with an etch resist 8, which remains omitted in areas to be demetalized. In these areas the metal layer 58 can then be removed by etching (Fig. 18A). In addition the area of the

surface relief 51 also initially remains left open here. In this embodiment the etch resist 8 is not removed and can take on additional decorative functions.

Next, a first metal layer 54 is applied by vapor deposition under vacuum (Fig. 18B). This first metal layer 54 covers, among other things, the surface relief 51. This first metal layer 54 is formed semi-transparent in this embodiment example.

Then the spacer layer 55 is now applied partially and registered relative to the surface relief 51 (Fig. 18C) and vapor-deposited over the whole surface with the second metal layer 56 (Fig. 16D). This second metal layer 56 is formed opaque. In areas which are not provided with the partial spacer layer 55, the second metal layer 56 lies directly on the first metal layer 54. An etch resist 81 is again printed onto the thus-generated second metal layer 56, which is opaque in this embodiment example, partially and registered (Fig. 18E).

The first 54 and the second 56 metal layers are then removed in one step in the left-open areas by etching (Fig. 18F). For this, the first 54 and the second 56 metal layers must consist of metals which can be removed with the same etching solution. Both metal layers preferably consist of the same metal, in particular preferably both metals consist of aluminum or copper.

The multilayer body 5 can now be applied to a banknote 7 again by means of an adhesive layer 57 (Fig. 18G). In this embodiment too, the substrate film 53 is removed (hot-embossing variant) and there is no window. The multilayer body 5 displays the desired effect in the case of direct observation from the side of the substrate film. Two demetalizing steps are enough for the method, for this

combined design with metallic areas with e.g. diffractive effects and areas with the color tilt effect.

5 A further method for producing a multilayer body with partial demetalization of the metal layers is represented in schematic – not to scale – side views in Figs. 19A to 19F.

Here too, a semi-transparent metal layer 54 is applied to the substrate film 53 with a surface relief 51 of the described type by vapor deposition under vacuum  
10 (Fig. 19A). Then the spacer layer 55 is now applied partially and registered relative to the surface relief 51 (Fig. 19B) and vapor-deposited over the whole surface with the second metal layer 56 (Fig. 19C). This second metal layer 56 is formed opaque. In areas which are not provided with the partial spacer layer 55, the second metal layer 56 lies directly on the first metal layer 54. Metallically  
15 diffractive effects are preferably present in these areas. An etch resist 8, which remains omitted in areas to be demetalized, is printed onto the second metal layer 56, which is opaque in this embodiment example, partially and registered (Fig. 19D). The first 54 and the second 56 metal layers are then removed in one step in the left-open areas by etching (Fig. 19E). For this, the first 54 and the  
20 second 56 metal layers must consist of metals which can be removed with the same etching solution. Both metal layers preferably consist of the same metal, in particular preferably both metals consist of aluminum or copper.

The multilayer body 5 can now be applied to a banknote 7 again by means of  
25 an adhesive layer 57 (Fig. 19F). In this embodiment too, the substrate film is removed (hot-embossing variant) and there is no window. The multilayer body 5 displays the desired effect in the case of direct observation from the side of the substrate film. Only one demetalizing step is enough for the method, for this

combined design with metallic areas with e.g. diffractive effects and areas with the color tilt effect.

5

10 Claims

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1. A method for producing a multilayer body according to a first group  
15 of steps a) to e) or a second group of steps a) to e), wherein the  
first group of steps comprises:
- a) providing a substrate film with a replication layer;
  - b) molding a surface relief appearing to the observer in the  
20 form of a three-dimensional free-form surface into a surface  
of the replication layer;
  - c) applying a first metal layer to the surface of the replication  
layer forming the surface relief;
  - d) wet-chemically applying an at least partially transparent  
25 spacer layer to the first metal layer, wherein the spacer layer  
forms a further surface relief, and wherein the surface relief  
and the further surface relief at least partially correlate; and
  - e) applying a second metal layer to the spacer layer,

wherein at least one of the two metal layers is formed semi-transparent; and

wherein the second group of steps a) to e) comprises:

- 5           a) providing a substrate film with a replication layer;
  - b) molding a surface relief appearing to the observer in the form of a three-dimensional free-form surface into a surface of the replication layer;
  - c) applying a first high-refractive-index dielectric layer to the surface of the replication layer forming the surface relief;
  - 10          d) wet-chemically applying an at least partially transparent spacer layer to the first dielectric layer, wherein the spacer layer forms a further surface relief, and wherein the surface relief and the further surface relief at least partially correlate; and
  - 15          e) applying a second high-refractive-index dielectric layer to the spacer layer; and
- wherein the spacer layer has a lower refractive index than the first and the second high-refractive-index layers.

- 20    2.       The method according to claim 1, wherein the surface relief appearing to the observer in the form of the three-dimensional free-form surface is formed by structures with a lens-like design generating one of a magnifying, a demagnifying and a distorting effect.

25

3. The method according to claim 1 or 2, wherein the spacer layer is applied by one of gravure printing, curtain coating, slot coating, spin coating and dip coating.
- 5 4. The method according to any one of claims 1 to 3, wherein the spacer layer is generated by application of a varnish.
5. The method according to claim 4, wherein the varnish is based on one of nitrocellulose, epoxy, polyester, rosin, acrylate, alkyd,  
10 melamine, PVA, PVC, isocyanate and urethane systems.
6. The method according to claim 4 or 5, wherein the varnish has a viscosity of from 5 mPa•s to 250 mPa•s.
- 15 7. The method according to claim 4 or 5, wherein the varnish has a viscosity of from 15 mPa•s to 200 mPa•s.
8. The method according to claim 4 or 5, wherein the varnish has a viscosity of from 20 and 170 mPa•s.
- 20 9. The method according to any one of claims 4 to 8, wherein the varnish has at least 30% by mass of a solvent with a polarity index greater than 3.0.

10. The method according to any one of claims 4 to 8, wherein the varnish has at least 50% by mass of a solvent with a polarity index greater than 3.0.
- 5 11. The method according to any one of claims 1 to 10, wherein the spacer layer is applied with a wet layer thickness of from 1  $\mu\text{m}$  to 20  $\mu\text{m}$ .
- 10 12. The method according to any one of claims 1 to 10, wherein, the spacer layer is applied with a wet layer thickness of from 2  $\mu\text{m}$  to 10  $\mu\text{m}$ .
13. The method according to any one of claims 1 to 12, wherein the spacer layer is dried after application.
- 15 14. The method according to claim 13, wherein the spacer layer is dried after application at a temperature of from 40°C to 200°C.
- 20 15. The method according to claim 13, wherein the spacer layer is dried after application at a temperature of from 40°C to 150°C.
- 25 16. The method according to any one of claims 1 to 15, wherein the one or both of the first metal layer and the second metal layer is generated by vacuum deposition of one of Cr, Al, Cu, Ti, Ni, Ag and Inconel.



17. The method according to claim 16, wherein the one or both of the first metal layer and the second metal layer is generated with a layer thickness of from 2 nm to 20 nm.
- 5
18. The method according to claim 16, wherein the one or both of the first metal layer and the second metal layer has a respective layer thickness of from 4 nm to 15 nm.
- 10 19. The method according to any one of claims 1 to 15, wherein the first and second high-refractive-index dielectric layers are generated by vacuum deposition of one of  $\text{TiO}_2$ ,  $\text{ZrO}_2$  and  $\text{ZnS}$ .
- 15 20. The method according to claim 19, wherein the first and second high-refractive-index dielectric layers have a respective layer thickness of from 10 nm to 200 nm.
- 20 21. The method according to any one of claims 1 to 20, wherein the first metal layer or second metal layer is generated by vacuum deposition of one of Al, Cr, Ag, and Cu, with a respective layer thickness of from 15 nm to 100 nm.
- 25 22. The method according to claim 21, wherein the first metal layer or the second metal layer is generated with a layer thickness of from 20 nm to 60 nm.

23. The method according to any one of claims 16 to 22, wherein the one or both of the first metal layer and the second metal layer are generated by vacuum coating or sputtering.
- 5
24. The method according to any one of claims 16 to 23, wherein the first and second high-refractive-index dielectric layers are generated by vacuum coating or sputtering.
- 10 25. The method according to any one claims 1 to 24, wherein the one or both of the first metal layer and the second metal layer is partially demetalized.
- 15 26. The method according to claim 25, wherein the one or both of the first metal layer and the second metal layer is partially demetalized by one or both of etching and lift-off.
- 20 27. The method according to claim 25 or 26, wherein the one or both of the first metal layer and the second metal layer is demetalized such that a demetalized area of the one or both of the first metal layer and the second metal layer forms a symbol, image, logo, alphanumeric character or a combination thereof.
- 25 28. The method according to any one of claims 25 to 27, wherein the first metal layer and the second metal layer are demetalized such

that the demetalized areas overlap in the viewing direction onto the surface normal of the plane spanned by the multilayer body.

29. The method according to any one of claims 25 to 28, wherein the  
5 first metal layer and the second metal layer are demetalized such that a metallic area of one of the first metal layer and the second metal layer overlaps the outer contour of a demetalized area of the respectively other of the first metal layer and the second metal layer.
- 10
30. A multilayer body, comprising:
- a substrate film;
  - a replication layer applied to a surface of the substrate film, into  
15 the surface of which facing away from the substrate film a surface relief appearing to the observer in the form of a three-dimensional free-form surface is introduced;
  - a first metal layer or a first high-refractive-index dielectric layer, which is arranged on the surface of the replication layer forming the surface relief;
  - 20 - a wet-chemically applied, at least partially transparent spacer layer, which is applied to a surface of the metal layer or of the high-refractive-index layer facing away from the replication layer, and which forms a further surface relief or a smooth surface, wherein the surface relief and the further surface relief at least  
25 partially correlate;
  - a second metal layer or a second high-refractive-index dielectric layer, which is applied to a surface of the spacer layer

facing away from the first metal layer or the first high-refractive-index dielectric layer.

31. The multilayer body of claim 30, wherein the surface relief  
5 appearing to the observer in the form of the three-dimensional free-form surface is comprises structures with a lens-like design generating one of a magnifying, a demagnifying and a distorting effect.
- 10 32. The multilayer body according to claim 30 or 31, wherein a structure depth of the further surface relief is at most 90% of a structure depth of the surface relief.
- 15 33. The multilayer body according to claim 30 or 31, wherein a structure depth of the further surface relief is at most 80% of a structure depth of the surface relief.
- 20 34. The multilayer body according to claim 30 or 31, wherein a structure depth of the further surface relief is at most 50% of a structure depth of the surface relief.
- 25 35. The multilayer body according to any one of claims 30 to 34, wherein the structure depth of the surface relief is 200 nm to 2000 nm.

36. The multilayer body according to any one of claims 30 to 34, wherein the structure depth of the surface relief is 300 nm to 1500 nm.
- 5 37. The multilayer body according to any one of claims 30 to 36, wherein the spacer layer has a dry layer thickness of from 200 nm to 800 nm, and wherein the dry layer thickness is variable over the spacer layer in the area of the surface relief appearing in the form of a three-dimensional free-form surface.
- 10 38. The multilayer body according to claim 37, wherein the dry layer thickness is from 300 nm to 700 nm.
- 15 39. The multilayer body according to any one of claims 30 to 38, wherein the spacer layer has a refractive index of from 1.35 to 1.65.
40. The multilayer body according to any one of claims 30 to 38, wherein the spacer layer has a refractive index of 1.5.
- 20 41. The multilayer body according to any one of claims 30 to 40, wherein the surface relief has microstructures with a depth-to-width ratio of less than 0.5.

42. The multilayer body according to any one of claims 30 to 40, wherein the surface relief has microstructures with a depth-to-width ratio of less than 0.4.
- 5 43. The multilayer body according to any one of claims 30 to 43, wherein the free-form surface is formed as a diffractive free-form element with a grating structure.
- 10 44. The multilayer body according to claim 43, wherein the grating structure comprises grating lines substantially following the outlines of the free-form surface, wherein the distance between the grating lines changes over the grating structure.
- 15 45. The multilayer body according to claim 44, wherein the distance between the grating lines changes continuously from the central area of the free-form surface out towards its edge.
- 20 46. The multilayer body according to any one of claims 43 to 45, wherein a period of the grating lines at least in a partial area of the grating structure is smaller than 50  $\mu\text{m}$ .
- 25 47. The multilayer body according to any one of claims 43 to 45, wherein a period of the grating lines at least in a partial area of the grating structure is smaller than 30  $\mu\text{m}$ .

48. The multilayer body according to any one of claims 43 to 45, wherein a period of the grating lines at least in a partial area of the grating structure is smaller than 15  $\mu\text{m}$ .
- 5 49. The multilayer body according to any one of claims 30 to 48, wherein the substrate film consists of a flexible plastic.
- 10 50. The multilayer body according to claim 49, wherein the flexible plastic comprises any of PET, PEN, MOPP, PP, PA, PC, COC and PVC.
- 15 51. The multilayer body according to any one of claims 30 to 50, wherein the substrate film has a layer thickness of from 4  $\mu\text{m}$  to 500  $\mu\text{m}$ .
52. The multilayer body according to any one of claims 30 to 50, wherein the substrate film has a layer thickness of from 8  $\mu\text{m}$  to 150  $\mu\text{m}$ .
- 20 53. The multilayer body according to any one of claims 30 to 50, wherein the substrate film has a layer thickness of from 12  $\mu\text{m}$  to 50  $\mu\text{m}$ .
- 25 54. The multilayer body according to any one of claims 30 to 53, wherein one of:

- i) one or both of the first meta layer and the second metal layer consists of Cr, Al, Cu, Ti, Ni, Ag or Inconel; and
- ii) the first and second high-refractive-index dielectric layers consist of TiO<sub>2</sub>, ZrO<sub>2</sub> or ZnS.

5

55. The multilayer body according to any one of claims 30 to 54, wherein one of:

- i) one or both of the first metal layer and the second metal layer has a layer thickness layer thickness of from 2 nm to 20 nm; and
- 10 ii) the first and second high-refractive-index dielectric layer has a layer thickness of from 10 nm to 200 nm.

56. The multilayer body according to any one of claims 30 to 54, wherein one of:

- 15 i) one or both of the first metal layer and the second metal layer has a layer thickness layer thickness of from 4 nm to 10 nm; and
- ii) the first and second high-refractive-index dielectric layer has a layer thickness of from 10 nm to 200 nm.

20 57. The multilayer body according to any one of claims 30 to 56, wherein the first metal layer or the second metal layer has a layer thickness of from 15 nm to 100 nm.

25 58. The multilayer body according to any one of claims 30 to 56, wherein the first metal layer or the second metal layer has a layer thickness of from 20 nm to 60 nm.



59. The multilayer body according to any one of claims 30 to 58, wherein the multilayer body has at least one varnish layer, which is dyed by means of one or both of a dye and a pigment.
- 5
60. The multilayer body according to claim 59, wherein the at least one varnish layer is arranged between the replication layer and the substrate film.
- 10 61. The multilayer body according to any one of claims 30 to 60, wherein the replication layer is dyed by means of one or both of a dye and a pigment.
- 15 62. The multilayer body according to any one of claims 30 to 61 obtained according to a method of any one of claims 1 to 29.
63. A security document with a multilayer body according to any one of claims 30 to 62.
- 20 64. The security document according to claim 63, wherein the security document is formed as any of an identification document, a visa document, a credit card, and a driver's license.

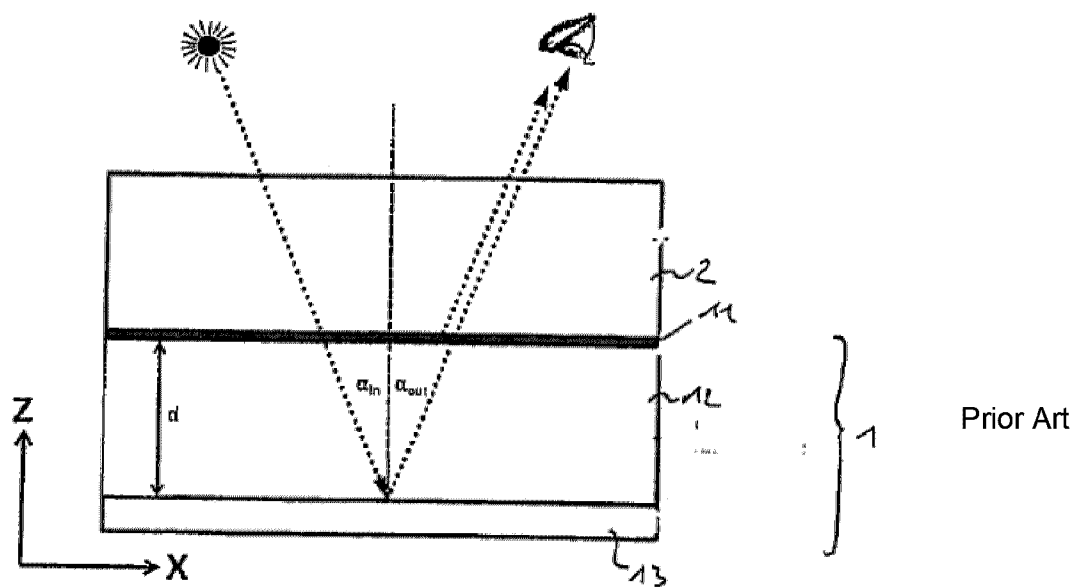


Fig. 1

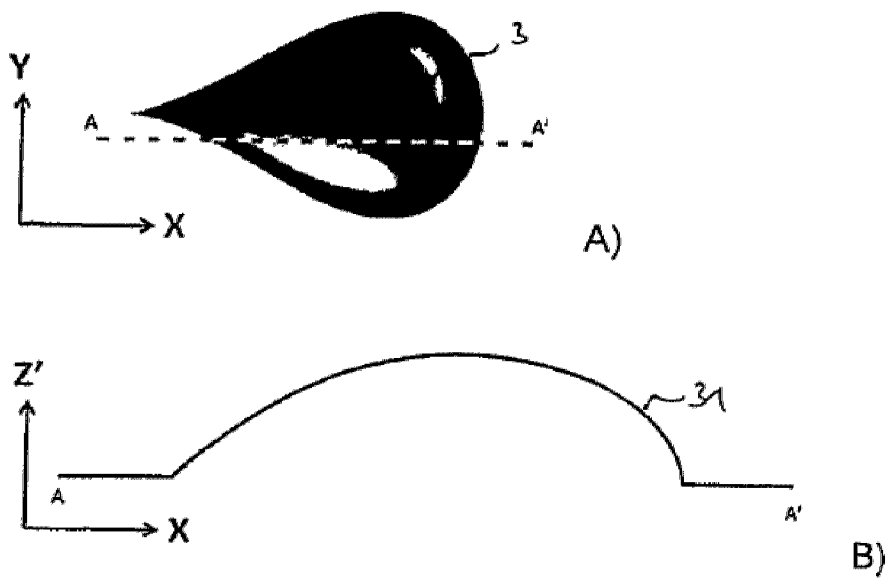


Fig. 2

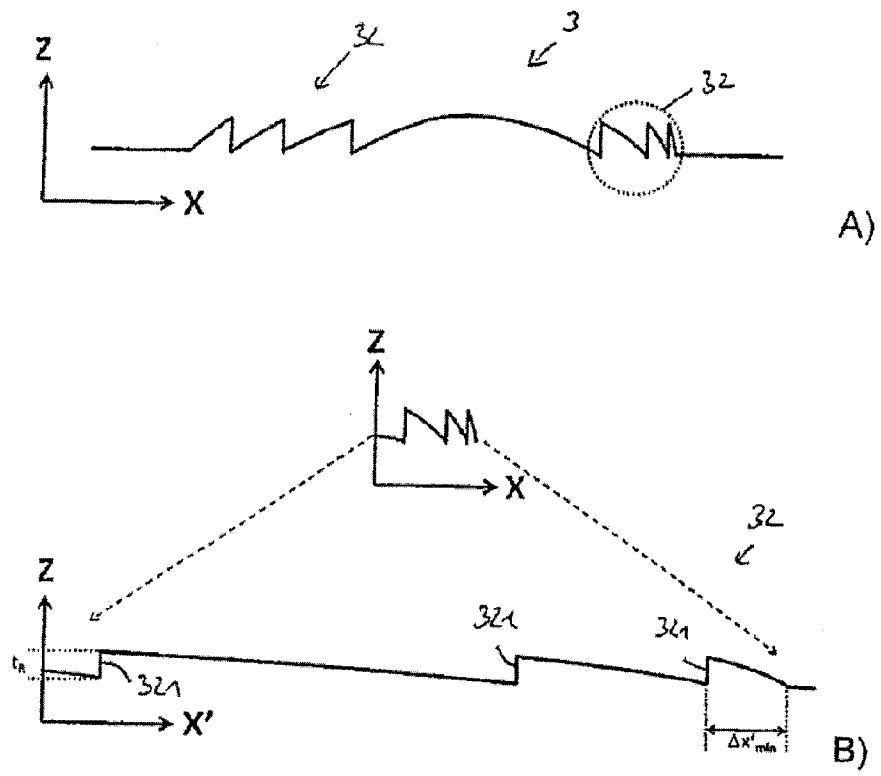


Fig. 3

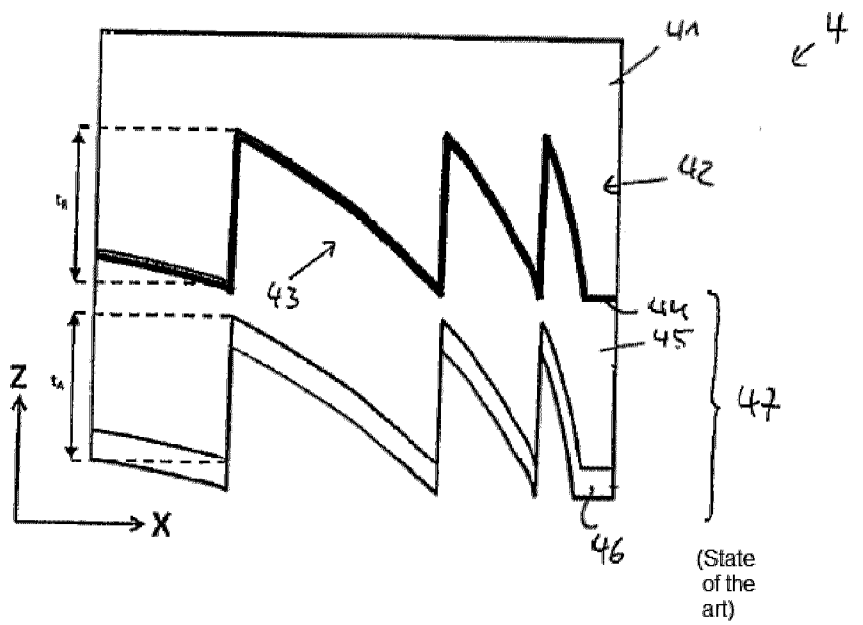


Fig. 4

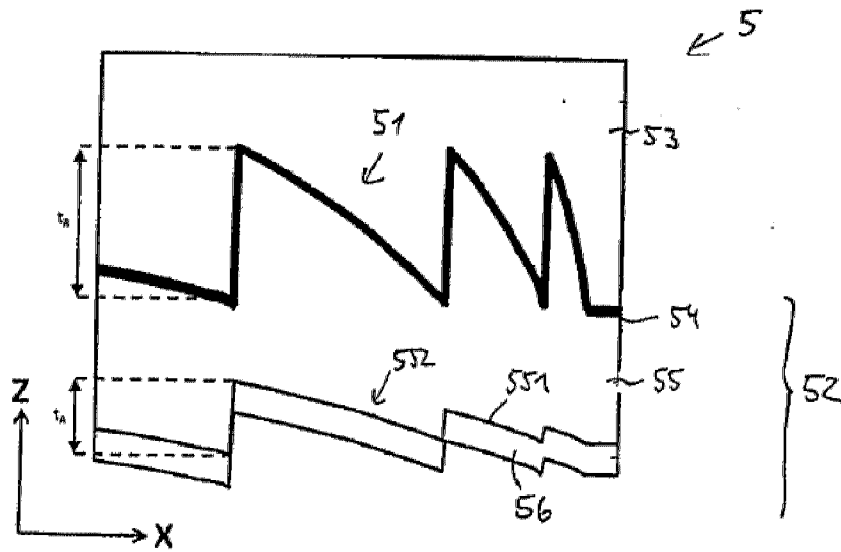


Fig. 5

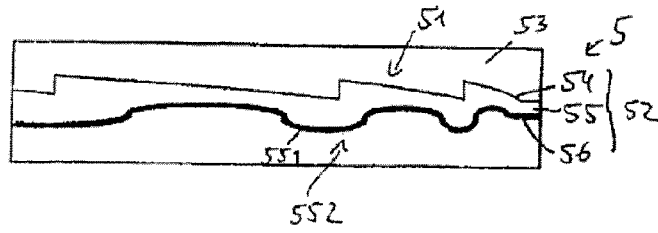


Fig. 6

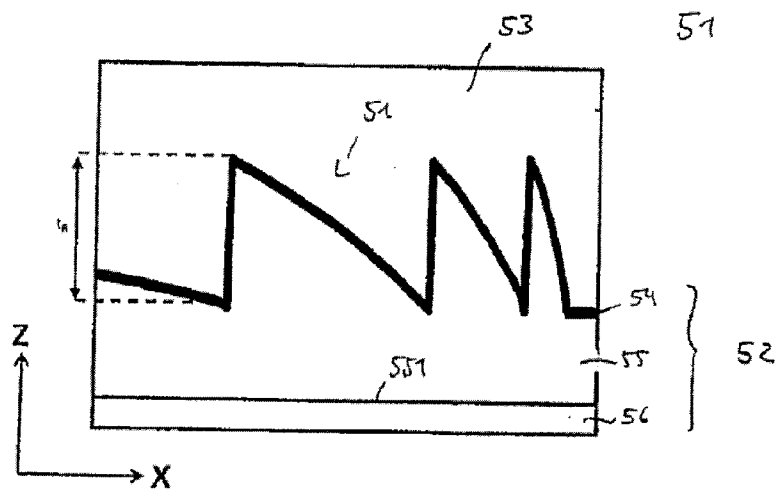


Fig. 7

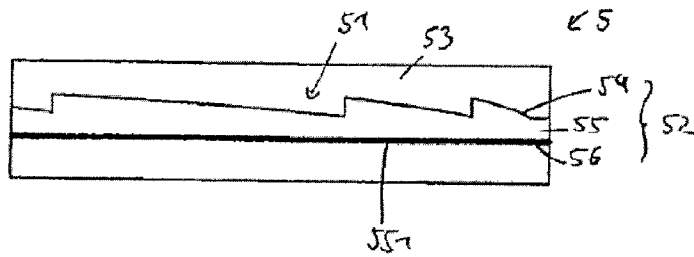


Fig. 8

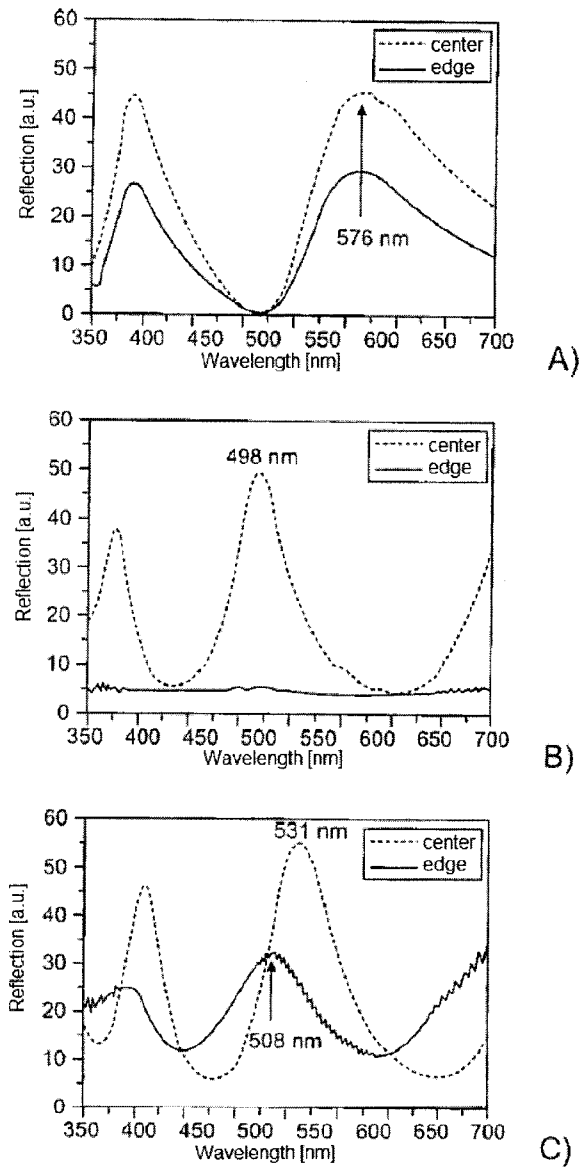


Fig. 9

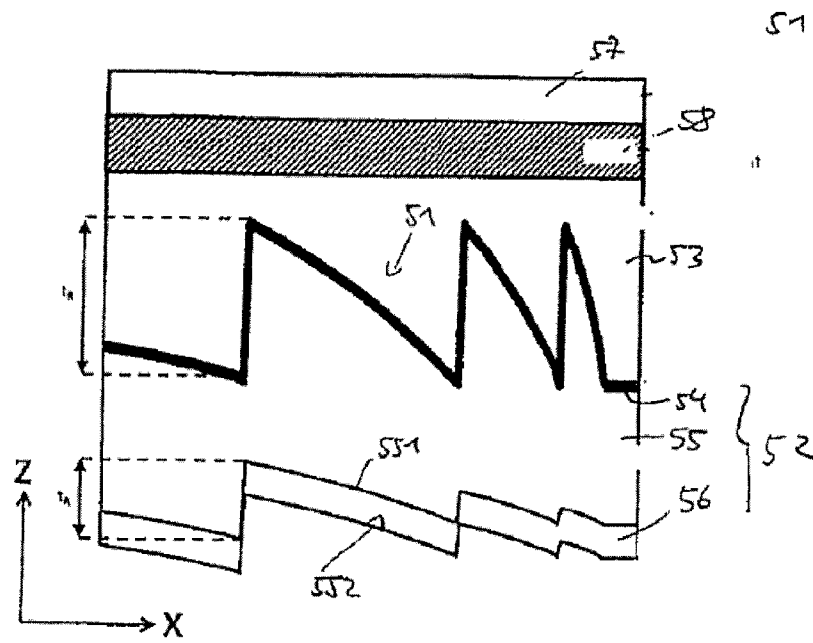


Fig. 10

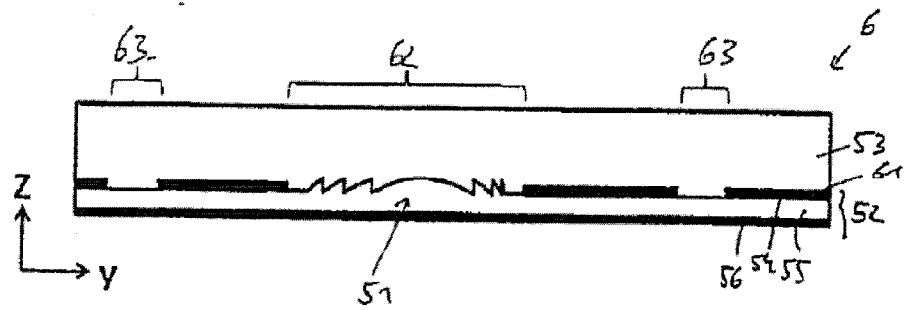


Fig. 11

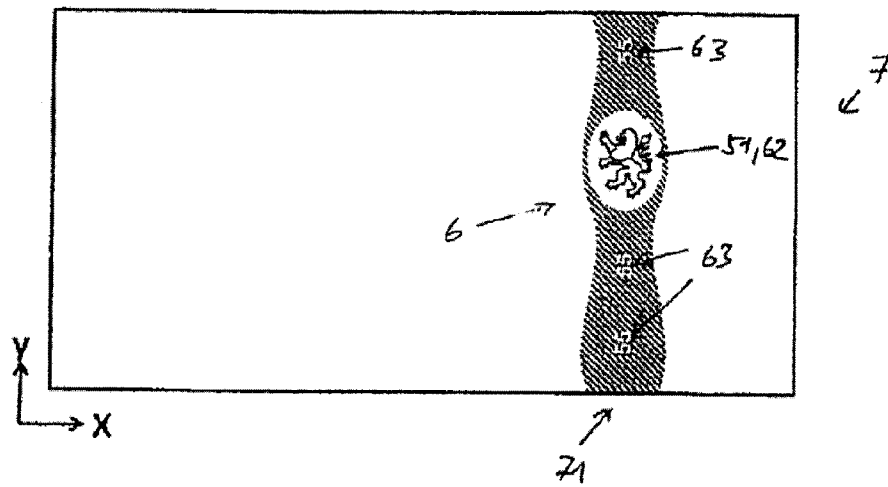


Fig. 12

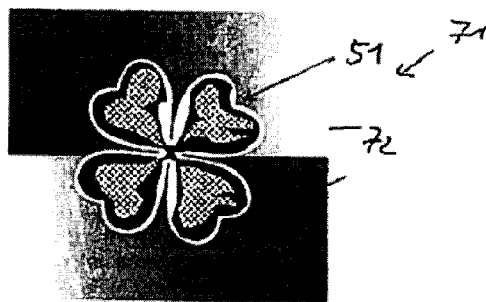
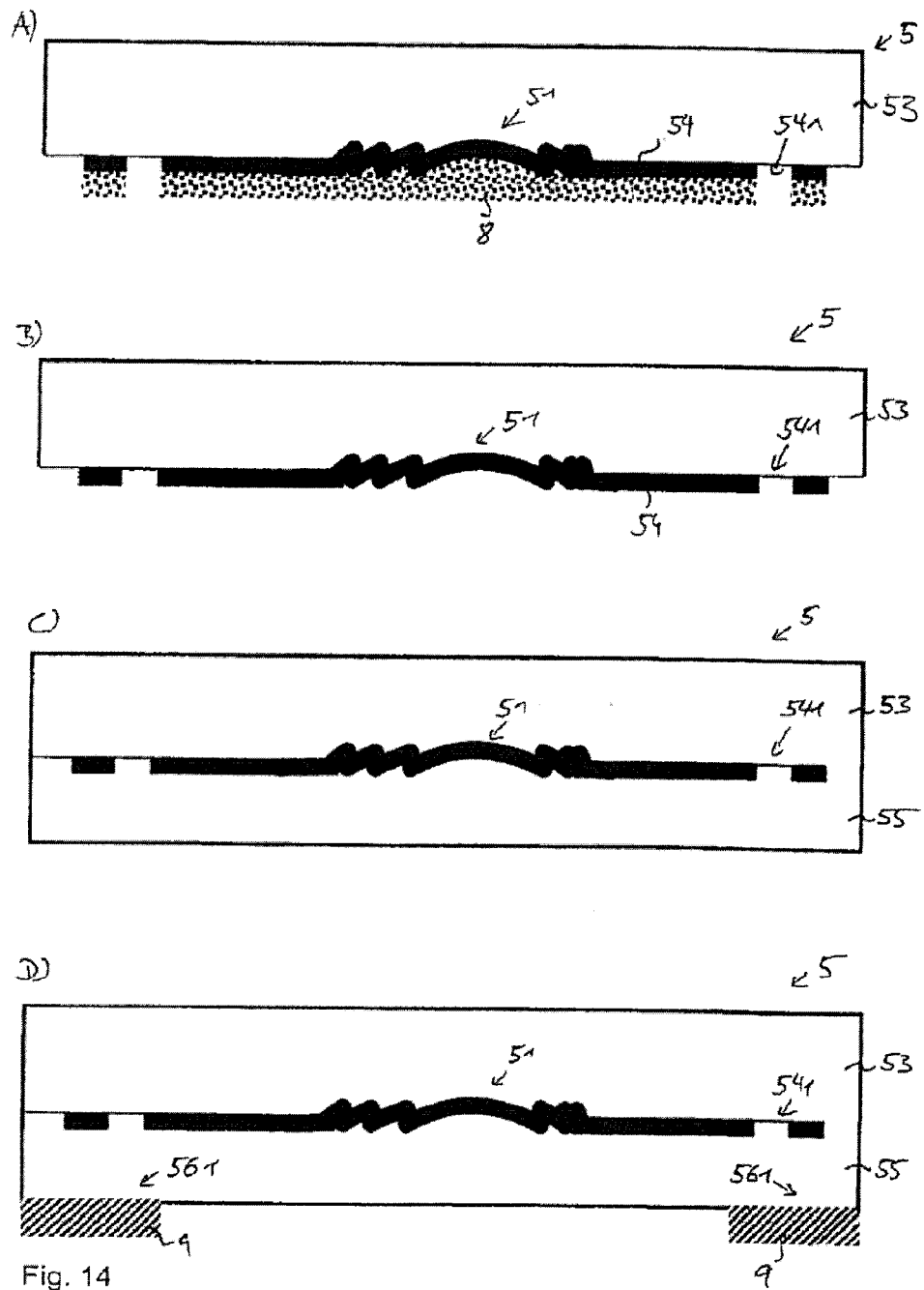


Fig. 13





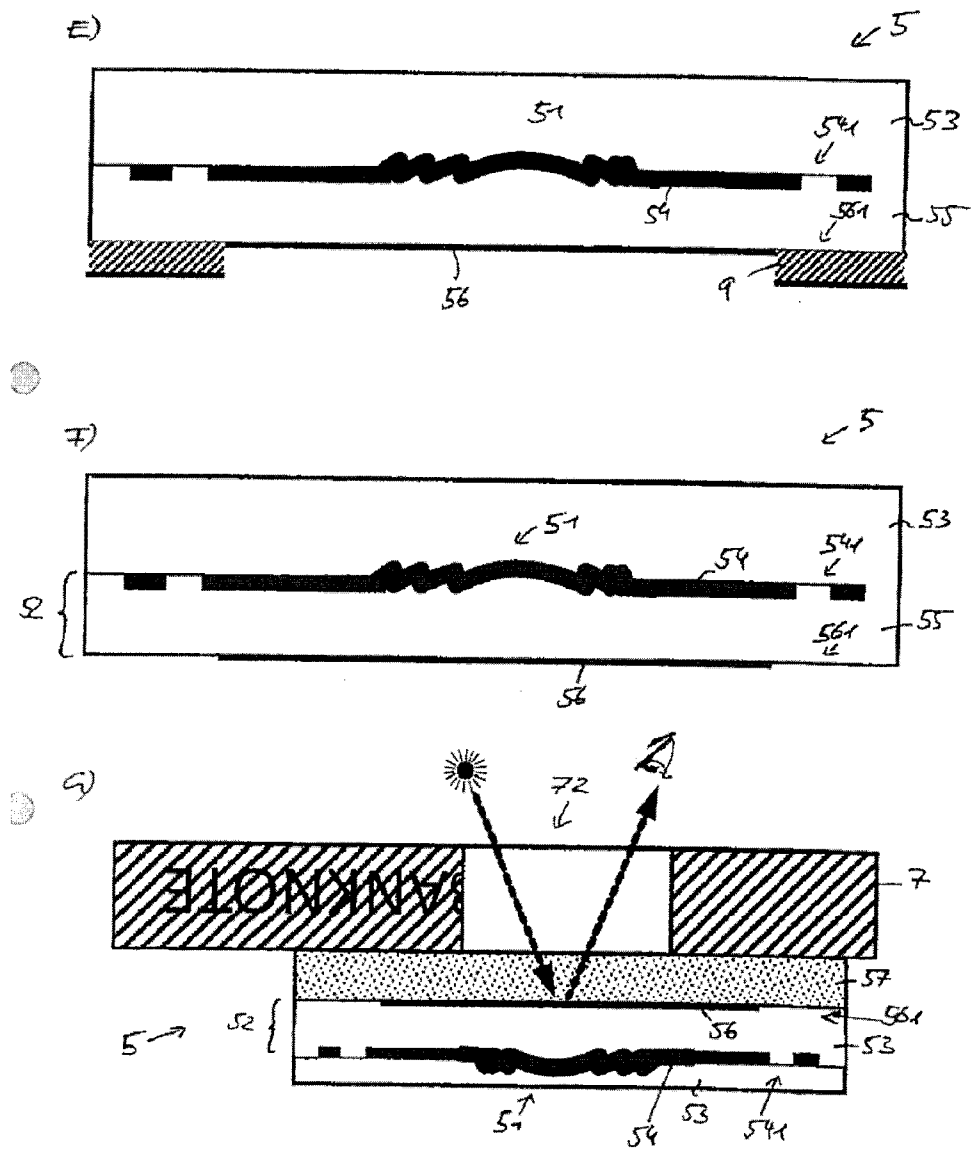


Fig. 14

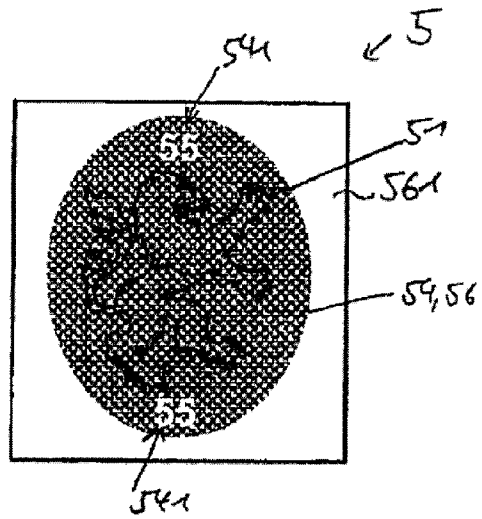


Fig. 15

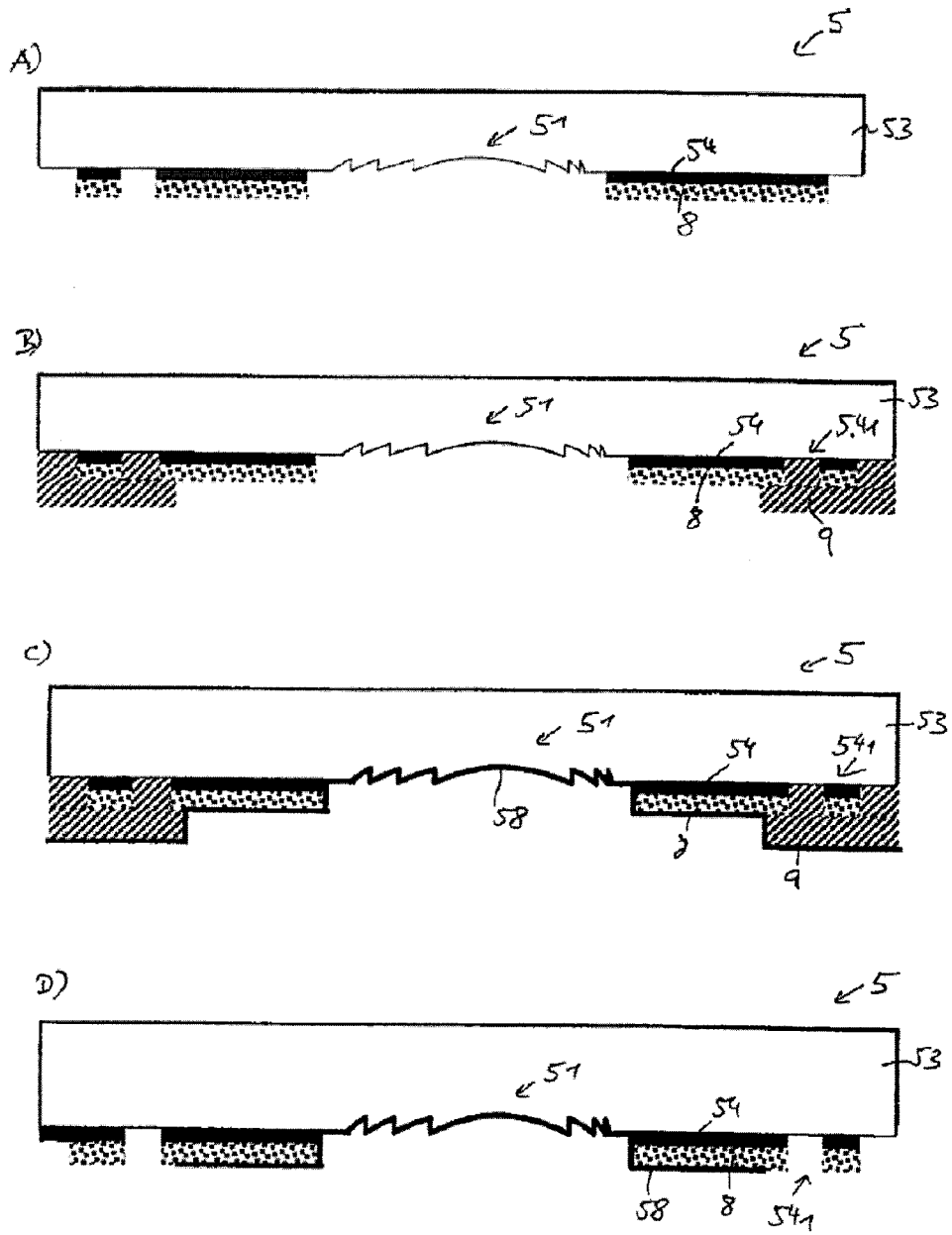


Fig. 16

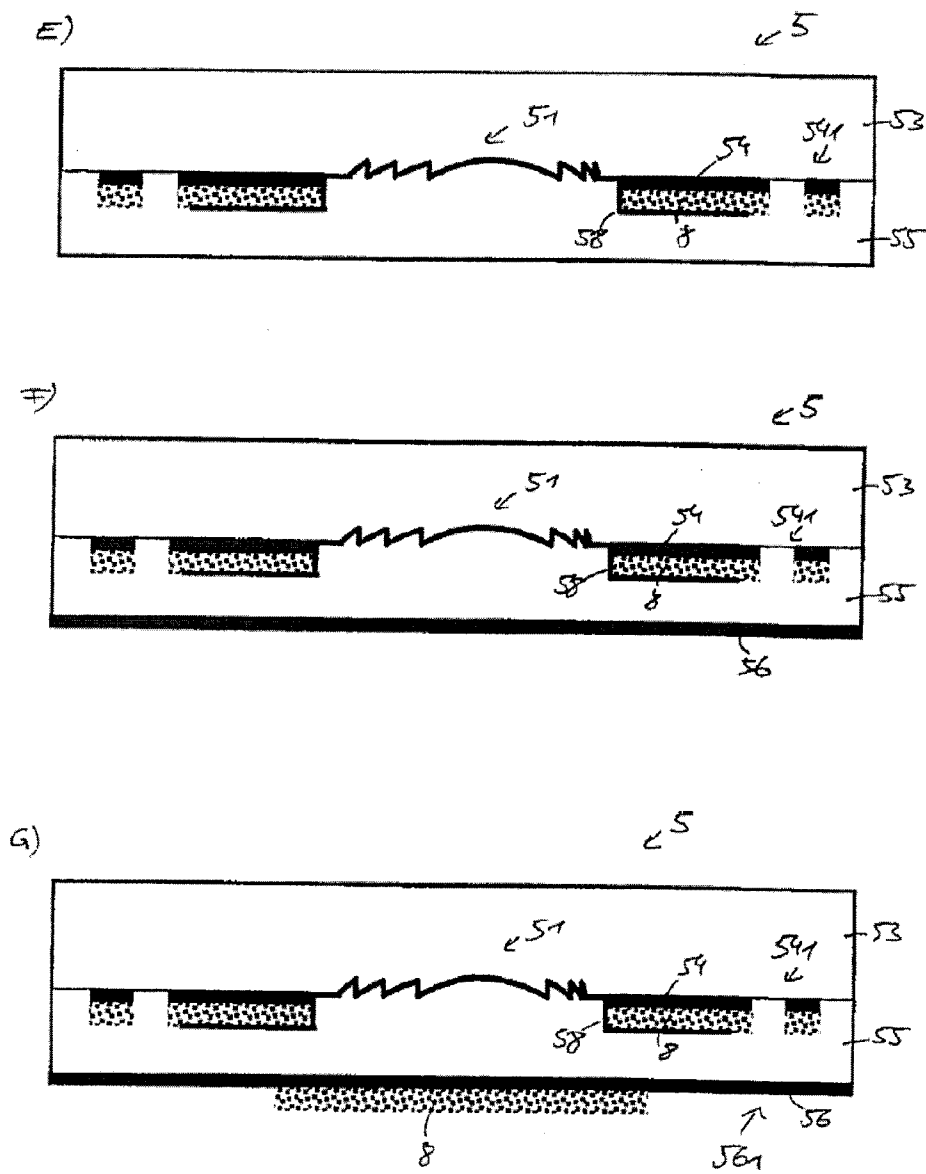


Fig. 16

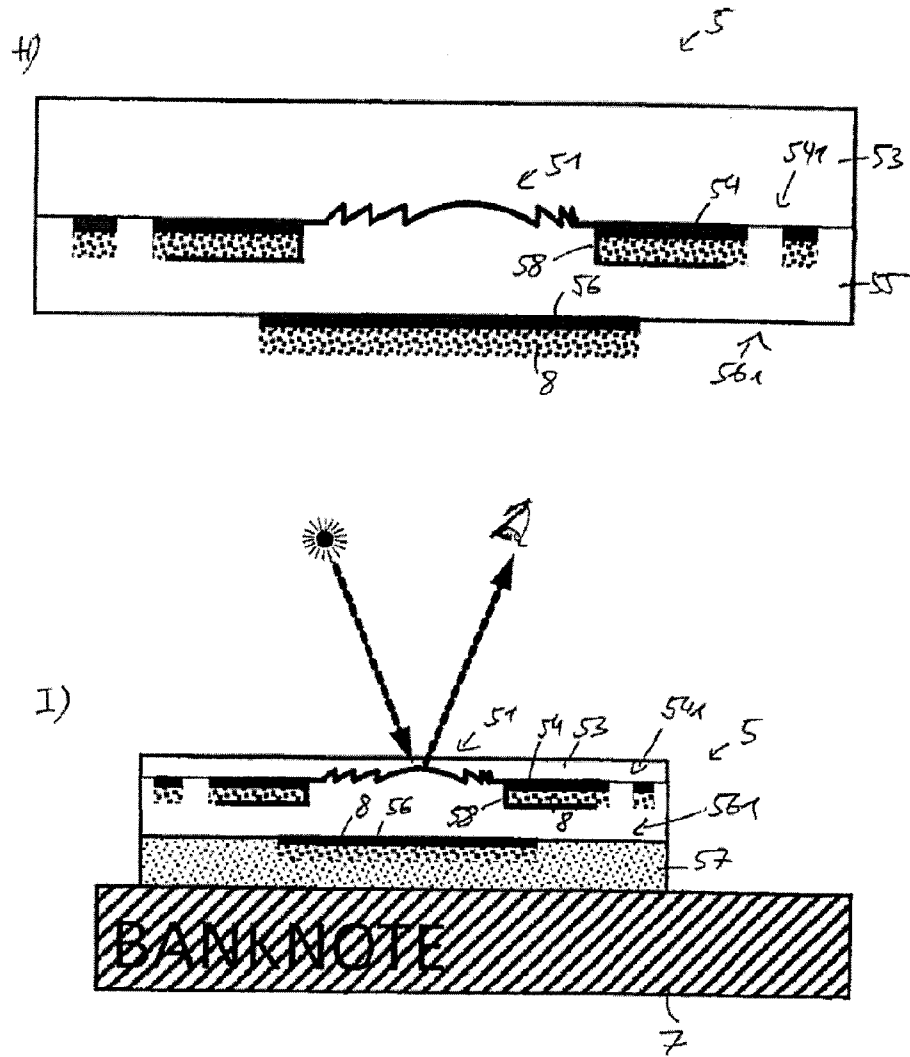


Fig. 16

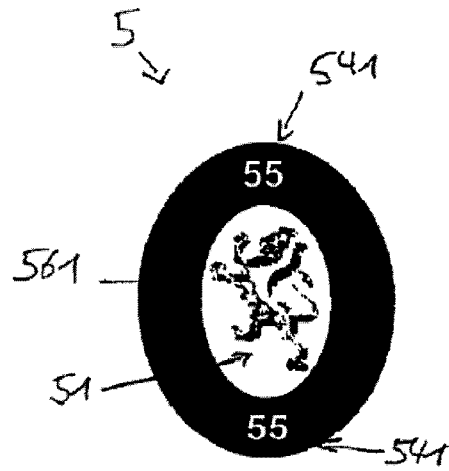


Fig. 17

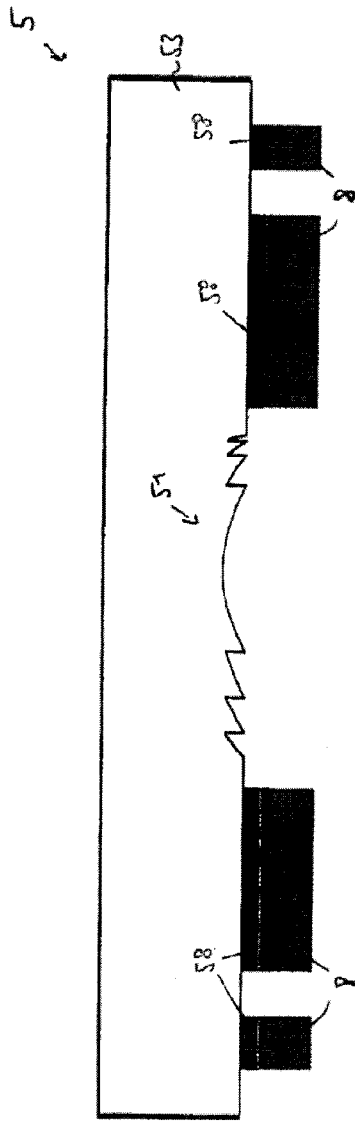


Fig. 18A



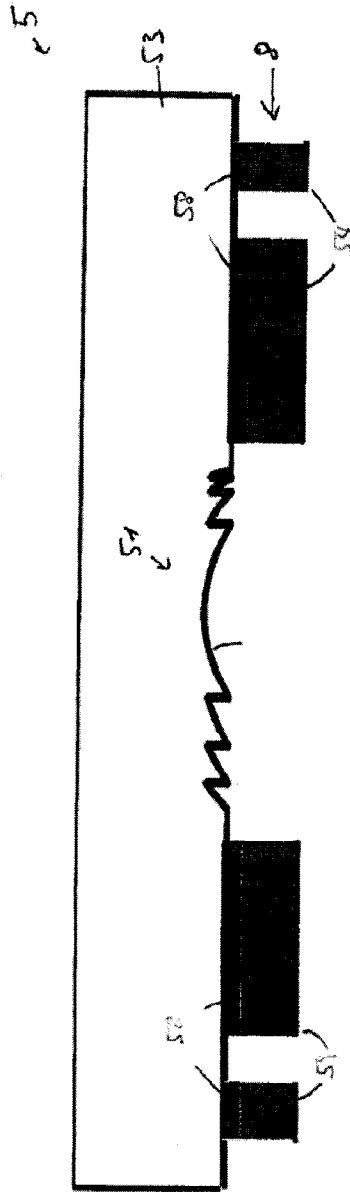


Fig. 18B

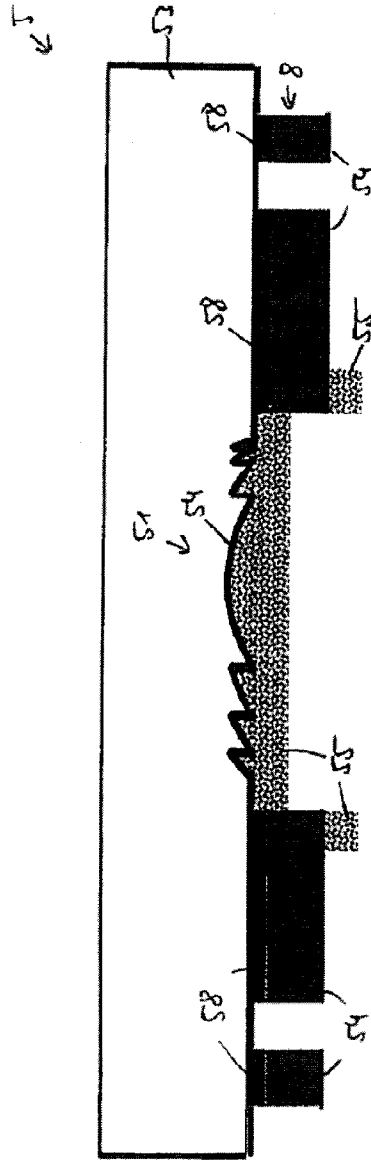


Fig. 18C

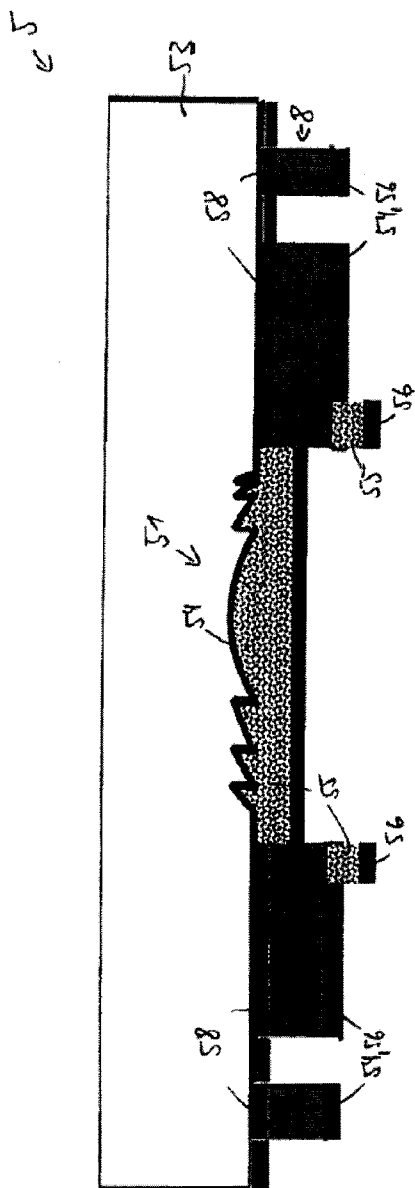


Fig. 18D

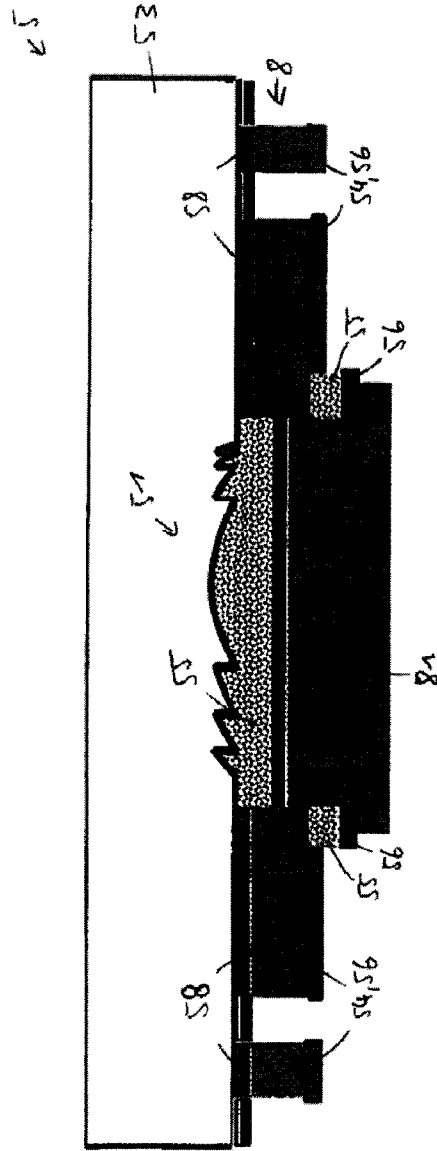


Fig. 18E

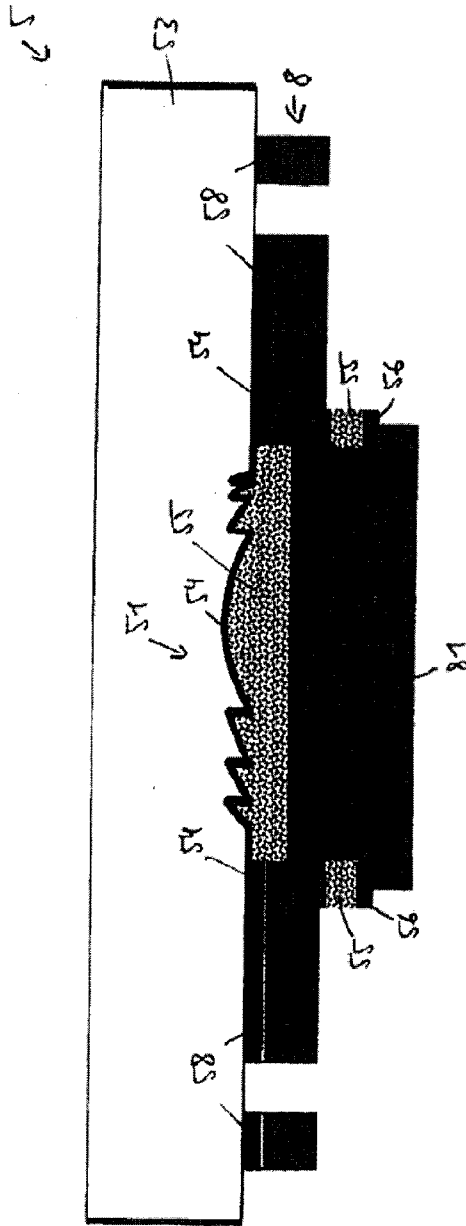


Fig. 18F

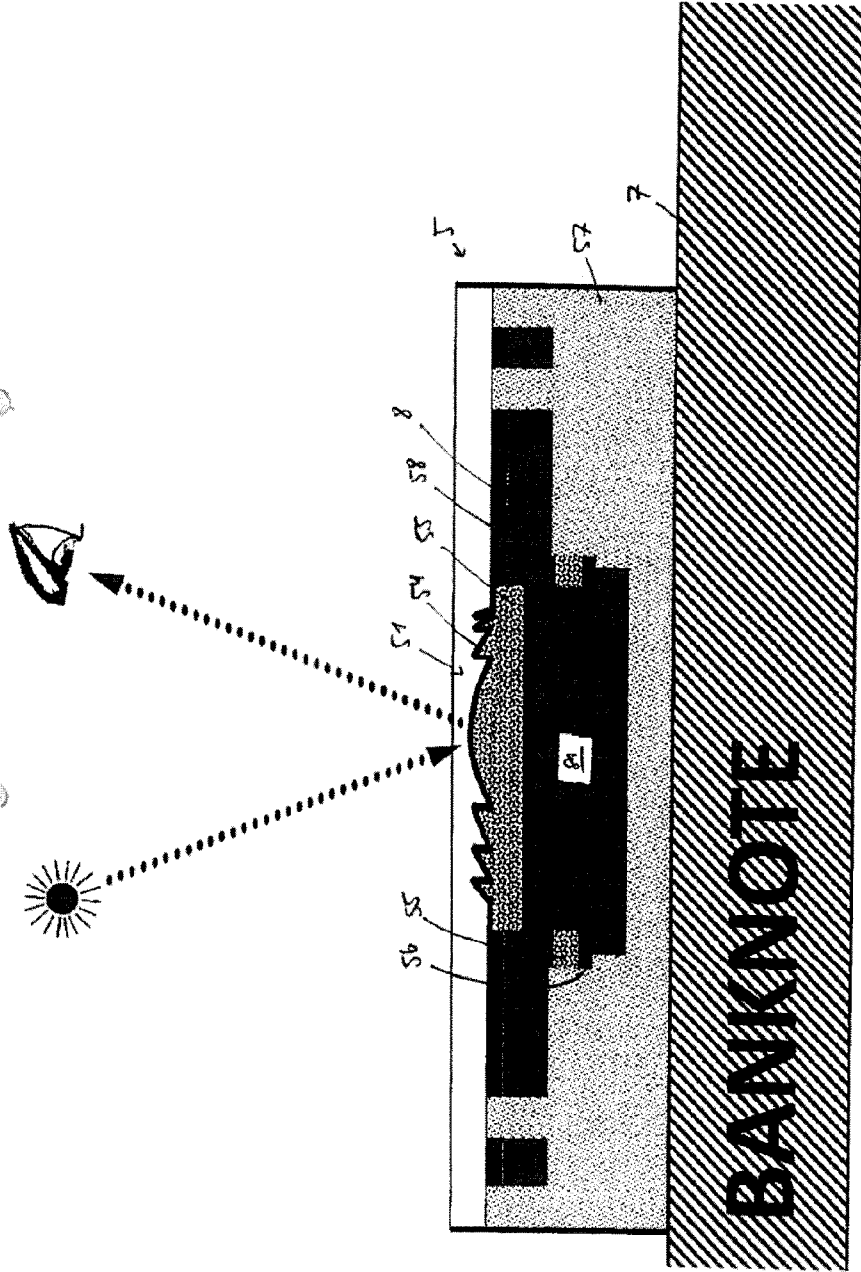


Fig. 18G

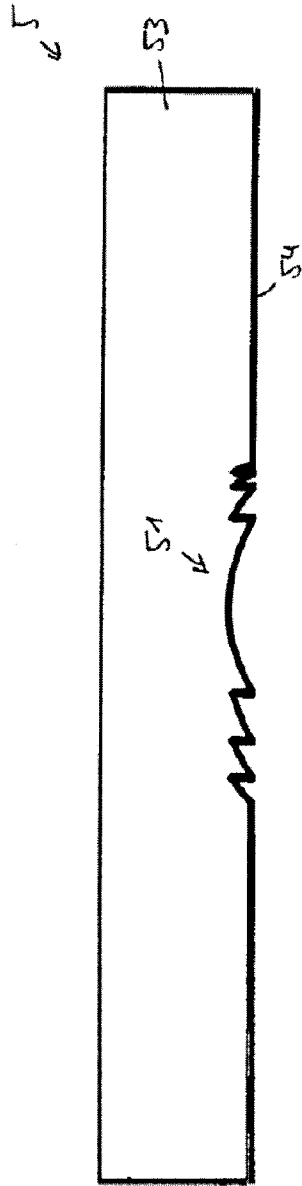


Fig. 19A

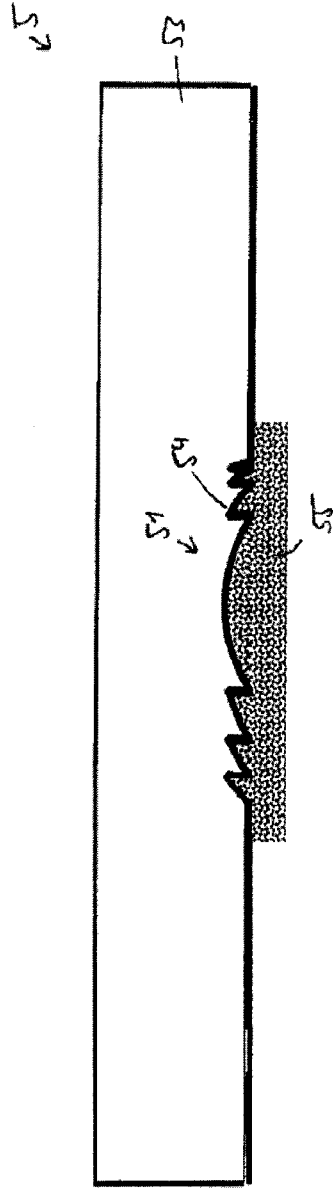


Fig. 19B



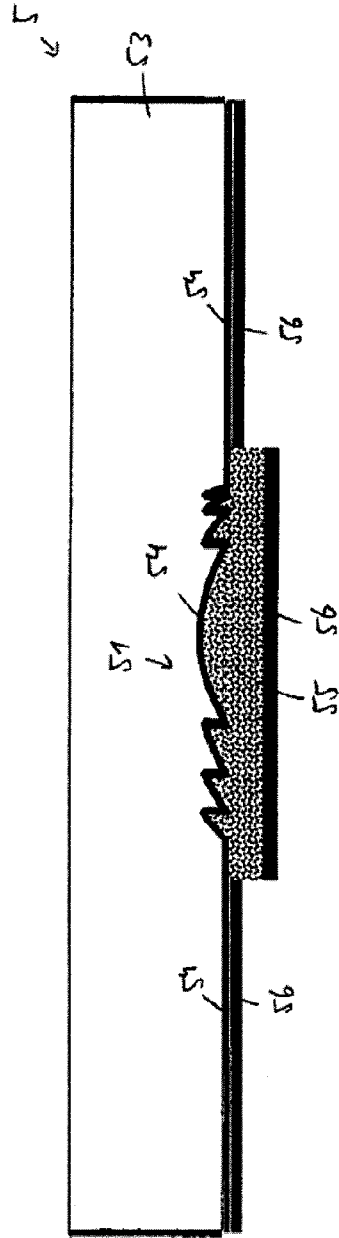


Fig. 19C

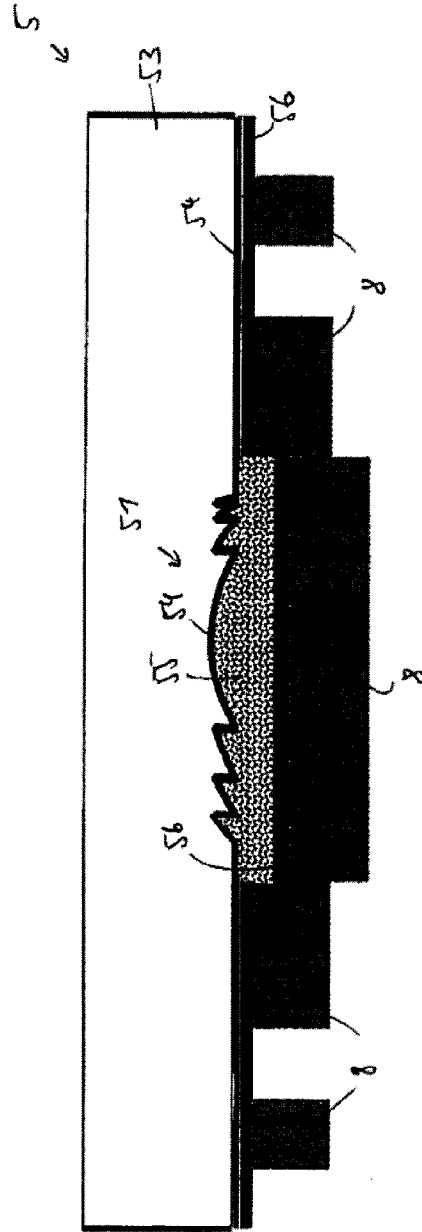


Fig. 19D

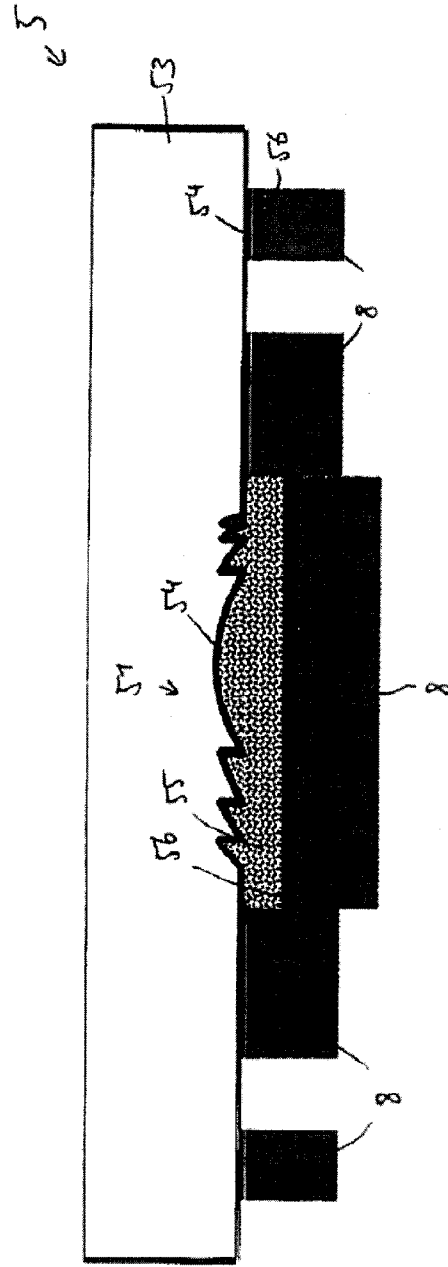


Fig. 19E

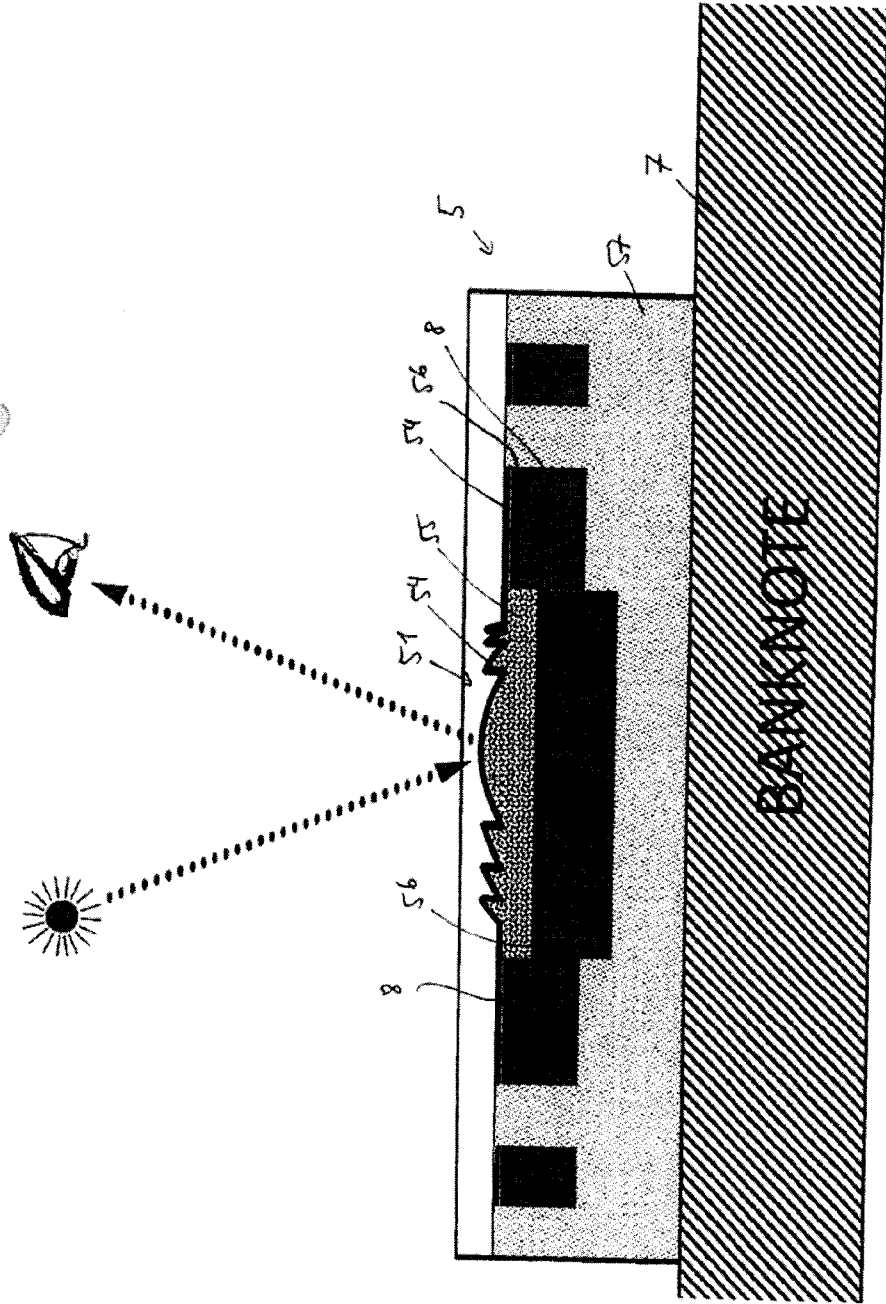


Fig. 19F

