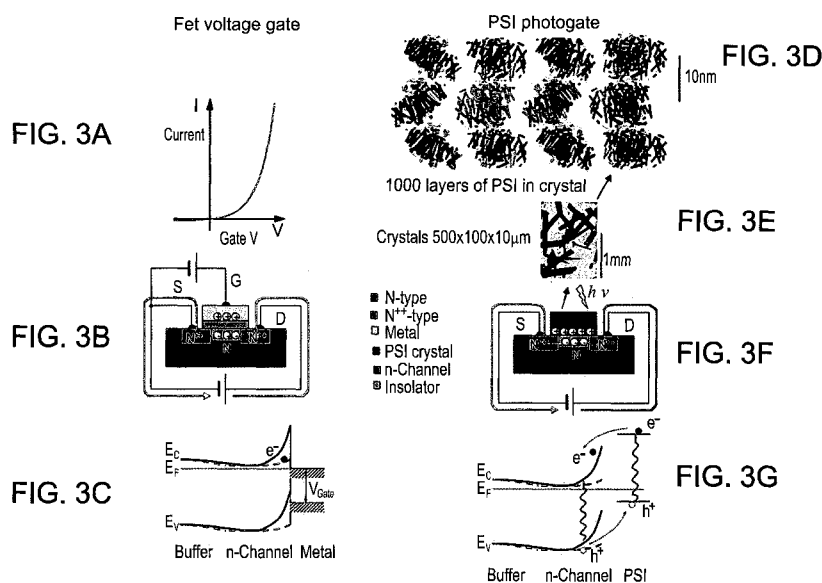




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(54) **Title:** CRYSTALLIZED PHOTOSYSTEM I UNITS FROM THE PEA PLANT AND THEIR USE IN SOLID STATE DEVICES



(57) **Abstract:** A crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I (PS I) is disclosed, wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all the individual photocatalytic activities. A method of generating same is also disclosed as well as uses of same.

CRYSTALLIZED PHOTOSYSTEM I UNITS FROM THE PEA PLANT AND THEIR USE IN SOLID STATE DEVICES

FIELD AND BACKGROUND OF THE INVENTION

5 The present invention relates to crystallized photocatalytic units and, more particularly, to solid supports fabricated with same. Some embodiments of the present invention relate to an optoelectronic device incorporating the photocatalytic units and method of fabricating same.

10 Green plants, cyanobacteria and photosynthetic bacteria capture and utilize sunlight by means of molecular electronic complexes, reaction centers that are embedded in their membranes. In oxygenic plants and cyanobacteria, photon capture and conversion of light energy into chemical energy take place in specialized membranes called thylakoids. The thylakoids are located in chloroplast in higher plants or consists of foldings of the cytoplasmic membrane in cyanobacteria. The thylakoids,
15 consisting of stacked membrane disks (called grana) and unstacked membrane disks (called stroma). The thylakoid membrane contains two key photosynthetic components, photosystem I and photosystem II, designated PS I and PS II, respectively. Photosynthesis requires PS II and PS I working in sequence, using water as the source of electrons and CO₂ as the terminal electron acceptor.

20 PS I is a transmembrane multisubunit protein-chlorophyll complex that mediates vectorial light-induced electron transfer. The nano-size dimension, an energy yield of approximately 58 % and the quantum efficiency of almost 1 [K. Brettel, Biochim.Biophys.Acta 1997, 1318 322-373] makes the reaction center a promising unit for applications in molecular nano-electronics. PS I mediates light-induced electron
25 transfer from plastocyanin or cytochrome C₅₅₃ to ferredoxin.

 The crystal structure of PS I from plant chloroplast was solved to 4.4 Å and then to 3.4 Å, [A. Ben Shem, et al., Nature 2003, 426 630-635; Amunts et al., Nature 2007, 447;58-63, both the references and their supplementary information being incorporated herein by reference]. Its intricate structure shows 12 core subunits, 4 different light-
30 harvesting membrane proteins (LHCI) assembled in a half-moon shape on one side of the core, 45 transmembrane helices, 168 chlorophylls, 3 Fe-S clusters and 2

phylloquinones. About 20 chlorophylls are positioned in strategic locations in the cleft between LHCI and the core.

WO2006/090381 and WO2008/023373 teach electronically active monolayers of genetically engineered cysteine mutants of PS I attached to solid surfaces.

5

SUMMARY OF THE INVENTION

According to an aspect of some embodiments of the present invention there is provided a crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I (PS I), wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all the individual photocatalytic activities.

According to some embodiments of the invention, the crystal further comprises a semi-conducting material in between the oriented layers.

According to some embodiments of the invention, the semi-conducting material comprises a plastic polymer.

According to some embodiments of the invention, the crystal further comprises at least one electron carrier in between the oriented layers

According to some embodiments of the invention, the electron carrier is a metal.

According to some embodiments of the invention, the crystal comprises a β angle of about 90.8 to 91.4°.

According to some embodiments of the invention, the crystal is characterized by having a space group of p21.

According to some embodiments of the invention, the crystal is characterized by unit cell dimensions of $a = 181.90 \pm 10 \text{ \AA}$, $b = 190.24 \pm 10 \text{ \AA}$ and $c = 219.66 \pm 10 \text{ \AA}$.

According to some embodiments of the invention, each of the layers comprises a photopotential of about 1V.

According to some embodiments of the invention, the crystal is capable of light energy conversion efficiency of approximately 47 %.

According to some embodiments of the invention, a surface of the crystal is attached to a bifunctional connecting molecule.

According to some embodiments of the invention, the surface of the crystal is covalently attached to the bifunctional connecting molecule.

According to some embodiments of the invention, the bifunctional connecting molecule is attached to a free carboxyl of the PS I.

5 According to some embodiments of the invention, the bifunctional connecting molecule is attached to a free primary amine of the PS I.

According to some embodiments of the invention, the bifunctional connecting molecule comprises a succinylimide moiety.

10 According to some embodiments of the invention, the crystal is capable of withstanding contact with a phosphate buffer for at least 10 minutes without dissolving.

According to an aspect of some embodiments of the present invention there is provided a method of crystallizing pea *Pisum sativum* PS I the method comprising:

(a) suspending a purified preparation of the PS I in a crystallization solution, the crystallization solution comprising:

- 15 (i) an aqueous buffer;
(ii) PEG 400;
(iii) a salt; and
(iv) PEG 6000 at a gradient of about 3.5-7 % w/v,
under conditions suitable to generate crystallized PS I;

20 (b) contacting the crystallized PS I with a protecting solution, the protecting solution comprising PEG 400 and PEG 6000,
thereby crystallizing the PS I.

According to some embodiments of the invention, a concentration of the PS I in the crystallization solution is about 9 mg/ml.

25 According to some embodiments of the invention, the salt comprises an ammonium salt.

According to some embodiments of the invention, the protecting solution further comprises a buffer and a salt.

30 According to some embodiments of the invention, the salt comprises an ammonium succinate salt.

According to some embodiments of the invention, the protecting solution further comprises a semiconducting material or a conducting material.

According to an aspect of some embodiments of the present invention there is provided a crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I (PS I), generated according to the method of the present invention.

5 According to an aspect of some embodiments of the present invention there is provided a composition, comprising a solid surface attached to a plurality of crystals of the present invention.

According to some embodiments of the invention, the crystallized PS Is are directly attached to the solid surface.

10 According to some embodiments of the invention, the directly attached is covalently attached.

According to an aspect of some embodiments of the present invention there is provided an optoelectronic device comprising at least one photocatalytic active crystal interposed between a first electrode and a second electrode, wherein the at least one photocatalytic active crystal is the crystal of the present invention.

15 According to an aspect of some embodiments of the present invention there is provided an optoelectronic device comprising a plurality of photocatalytic active crystals interposed between a first electrode and a second electrode, wherein at least one of the photocatalytic active crystals is the crystal of the present invention.

20 According to an aspect of some embodiments of the present invention there is provided a circuitry, comprising an arrangement of optoelectronic devices, wherein at least one of the optoelectronic devices is the optoelectronic device of the present invention and wherein at least two of the optoelectronic devices are in electrical communication.

25 According to some embodiments of the invention, the device or circuitry serve as a component in a photodiode, a phototransistor, a photogate, a logic gate, a solar cell or an optocoupler.

According to some embodiments of the invention, at least one of the first electrode and the second electrode is transparent to light capable of activating the Photosystem.

30 According to some embodiments of the invention, the device or circuitry further comprise a cavity having a base and walls formed in an isolating substrate, wherein the

first electrode is deposited onto the base, and the second electrode is supported by the walls.

According to an aspect of some embodiments of the present invention there is provided a transistor, comprising a substrate formed with a source region and a drain region being laterally displaced from the source region, and a photogate attached to a surface of the substrate at least between the regions, wherein the photogate comprises the crystal of the present invention.

According to an aspect of some embodiments of the present invention there is provided a method of fabricating an optoelectronic device, comprising depositing the crystal of the present invention on a first electrode and depositing a second electrode on the crystal.

According to some embodiments of the invention, the method further comprises passivating the first electrode prior to the deposition of the crystal, so as to allow binding of the crystal to the first electrode.

According to some embodiments of the invention, the passivation of the first electrode comprises attaching bifunctional connecting molecules on a surface of the first electrode.

According to some embodiments of the invention, the bifunctional connecting molecules form a sulfide bond with the first electrode and an amide bond with the crystal.

According to some embodiments of the invention, the method further comprises coating the crystal with a protective coat prior to the deposition of the second electrode.

According to some embodiments of the invention, the method further comprises forming a cavity having a base and walls in an isolating substrate and depositing the first electrode onto the base, wherein the deposition of the second electrode is done such that the second electrode is supported by the walls.

According to an aspect of some embodiments of the present invention there is provided a method of forming a transistor, comprising electrically coupling the crystal of the present invention to at least an exposed surface of a substrate between a source region formed in the substrate and a drain region formed in the substrate, such that photo induced charge separation in the crystal results in formation of a conducting

channel region between the source region and the drain region, thereby forming the transistor.

According to some embodiments of the invention, the method further comprises modifying the surface prior to the deposition of the crystal, so as to allow binding of the crystal to the surface.

According to some embodiments of the invention, the modifying comprises chemisorption of surface modifying molecules which comprise free amine groups.

According to some embodiments of the invention, the surface modifying molecules bind directly to the crystal.

According to some embodiments of the invention, the method further comprises contacting the modified surface with connecting molecules such as to form an amide bond between the surface modifying molecules and the connecting molecules.

According to some embodiments of the invention, the method further comprises generating conditions for binding of the connecting molecules to the crystal.

According to some embodiments of the invention, the method further comprises forming an isolating layer on the surface prior to the deposition of the crystal, so as to allow binding of the crystal to the isolating layer.

Unless otherwise defined, all technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which this invention belongs. Although methods and materials similar or equivalent to those described herein can be used in the practice or testing of the present invention, suitable methods and materials are described below. All publications, patent applications, patents, and other references mentioned herein are incorporated by reference in their entirety. In case of conflict, the patent specification, including definitions, will control. In addition, the materials, methods, and examples are illustrative only and not intended to be limiting.

BRIEF DESCRIPTION OF THE DRAWINGS

The invention is herein described, by way of example only, with reference to the accompanying drawings. With specific reference now to the drawings in detail, it is stressed that the particulars shown are by way of example and for purposes of

illustrative discussion of the preferred embodiments of the present invention only, and are presented in the cause of providing what is believed to be the most useful and readily understood description of the principles and conceptual aspects of the invention. In this regard, no attempt is made to show structural details of the invention in more detail than is necessary for a fundamental understanding of the invention, the description taken with the drawings making apparent to those skilled in the art how the several forms of the invention may be embodied in practice.

In the drawings:

FIGs. 1A-C are schematic representations of PS I and PS I attached to a solid surface. Figure 1A illustrates the molecular structure of PS I and Light-induced charge separation across the electron transport chain. Figures 1B-C are schematic presentations of a PS I monolayer on GaAs attached by the chemisorption of linker molecules;

FIG. 2 comprises symbols representing the electronic circuit of gated field effect transistor (FET). The line indicates n channel with the source and drain leaving it at right angles. Gate voltage (arrow) in a n channel junction gate FET (JFET) (left) and light induce voltage by (two arrows) in a PS I crystal photogate of FET (right);

FIGs. 3A-G are schematic representations of a voltage gate and a PS I crystal photogate of a FET transistor. Gate voltage exponentially increase current (Figure 3A) between drain and source in metal oxide semiconductor FET (MOSFET) transistors (Figure 3B) by decreasing band banding in the n++/n/n++ junction (Figure 3C). The serially oriented multilayers of PS I (Figure 3D) in the crystals (Figure 3E) serve as photogate over the n channel (Figure 3F) that open the channel by decreasing band banding (Figure 3G);

FIG. 4 is a schematic representation of an electronic equivalent circuit of a photovoltaic cell. Such a device (left) can be described as comprising a diode dark current (R_{dark}), photocurrent (J_{sc}) going through a shunt (R_{sh}) and a series resistor (R_s). A symbol representing the electronic circuit of the photovoltaic cell is shown on the right;

FIG. 5A is a schematic illustrating how the PS I crystals bind to a solid surface. A monolayer of bifunctional connecting molecules are fabricated on the Au surface

through sulfide bonding and the PS I crystals protein amines are bound to them by reacting with the succinylimide moiety;

FIG. 5B is a schematic presentation of a PS I crystal millimetric photovoltaic cell. Light induced charge separation in PS I crystals drives current in a solid state scaffold consisting of a bottom metal electrode inside a millimeter-cavity drilled in a Si_3N_4 (gray) insulating layer topped by a conductive transparent layer such as InSnO2 (ITO) electrode;

FIG. 6 is a schematic illustration of an optoelectronic device, according to various exemplary embodiments of the present invention;

FIG. 7 is a schematic illustration of a photogate or light sensor device, according to various exemplary embodiments of the present invention;

FIGs. 8A-B are schematic illustrations of a field effect transistor (FET) device, according to various exemplary embodiments of the present invention;

FIG. 9 is a flowchart diagram of a method suitable for fabricating an optoelectronic device, according to various exemplary embodiments of the present invention;

FIG. 10 is a flowchart diagram of a method suitable for fabricating a transistor, according to various exemplary embodiments of the present invention;

FIGs. 11A-B are graphs illustrating the photopotential generated in PS I crystals bound to gold electrode by succinimide chemistry. (Figure 11A) A positive photopotential is generated by crystals bound through the oxidizing end of PS I and (Figure 11B) a negative photopotential is generated by crystals bound through the reducing end of PS I;

FIG. 12A is a graph illustrating the photopotential generated by oriented PS I crystals bound to n-GaAs surface by succinimide chemistry;

FIG. 12B is a schematic representation of n-GaAs;

FIGs. 12C-D are schematic representations illustrating the oriented binding of PS I crystals to GaAs surface using succinimide chemistry; and

FIGs. 13A-B are graphs comparing the photopotential generated by the PS I crystals of the present invention bound to gold-plated glass (Figure 13A) compared to previously known PS I crystals bound to gold-plated glass (Figure 13B).

DESCRIPTION OF SPECIFIC EMBODIMENTS OF THE INVENTION

The present invention, in some embodiments thereof, relates to crystallized photocatalytic units and, more particularly, to solid supports fabricated with same. Some embodiments of the present invention relate to an optoelectronic device
5 incorporating the photocatalytic units and method of fabricating same.

Before explaining at least one embodiment of the invention in detail, it is to be understood that the invention is not necessarily limited in its application to the details set forth in the following description or exemplified by the Examples. The invention is capable of other embodiments or of being practiced or carried out in various ways.

10 Photosynthesis is the biological process that converts electromagnetic energy into chemical energy through light and dark reactions. In oxygenic plants and cyanobacteria, photon capture and conversion of light energy into chemical energy take place in specialized membranes called thylakoids. In higher plants, the thylakoids are located in the chloroplast.

15 PS I is a transmembrane multisubunit protein-chlorophyll complex that mediates vectorial light-induced electron transfer from plastocyanin or cytochrome C₅₅₃ to ferredoxin. The nano-size dimension, an energy yield of approximately 58 % and the high quantum efficiency makes the reaction center a promising unit for applications in molecular nano-electronics.

20 Crystallization of PS I is known in the art – see for example A. Ben Shem, et al., *Nature* 2003, 426 630-635; Amunts et al., *Nature* 2007, 447;58-63. However, until presently no crystal has been generated such that the proteins within are stacked in such a way that they are all capable of absorbing visible light in an efficient manner and converting the energy into electric potential and current.

25 While reducing the present invention to practice, the present inventors have uncovered a novel way of crystallizing pea *Pisum sativum* PS I. Using very precise crystallization methods, the present inventors generated a crystal whereby the dry membrane proteins within retain their capacity to generate a photo-potential of approximately 1 V/ layer. The novel crystal structure enables generation of photo-
30 potential over 20 V/ 5 μ m crystal width that potentially may increase to up to 500 V.

Thus, according to one aspect of the present invention, there is provided a crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I

(PS I), wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all individual photocatalytic activities.

As used herein, the phrase "pea *Pisum sativum*" refers to the plant of Taxonomy
5 ID: 3888.

The phrase "Photosystem I" as used herein, refers to the monomeric, protein-chlorophyll complex which serves as a photocatalytic unit within the thylakoid membrane of plants.

The PS I complex typically comprises chlorophyll molecules which serve as
10 antennae which absorb photons and transfer the photon energy to P700, where this energy is captured and utilized to drive photochemical reactions. It is ellipsoidal in shape and has dimensions of about 9 by 15 nanometers.

In addition to the P700 and the antenna chlorophylls, the PS I complex contains a number of electron acceptors. An electron released from P700 is transferred to a
15 terminal acceptor at the reducing end of PS I through intermediate acceptors, and the electron is transported across the thylakoid membrane. For more information on the composition of PS I in the pea plant see for example [A. Ben Shem, et al., *Nature* 2003, 426 630-635], incorporated herein by reference.

The term "crystal", as used herein, refers to a three dimensional ordered
20 arrangement of atoms or molecules, which possesses symmetry characteristics. The ordering of the atoms or molecules is manifested by an elementary lattice unit, (also known as a "unit cell") having definite faces that intersect at definite angles, and possesses one or more symmetry characteristics which are described mathematically by a symmetry group (also known as the "crystallographic point group"). The overall
25 structure of the crystal is periodic, namely, it possesses a translational symmetry, and the elementary lattice unit defines the periodicity of the crystal. The translational symmetry of the crystal can extend over at least 10^3 or at least 10^4 or at least 10^5 or at least 10^6 elementary lattice units. The symmetry group that describes the symmetry characteristics of the crystal is referred to as a "space group", and is defined as the
30 combination of the symmetry group that describes the translational symmetry with the crystallographic point group. A crystal and its space group can be experimentally

identified by means of X-ray crystallography. All these are well known to those skilled in the art of crystallography.

According to this aspect of the present invention the crystal comprises ordered layers of PS I, each one being stacked above the other such that the PS Is are physically and electronically coupled. The crystallization is such that the oxidizing end of a PS I is layered above the reducing end of the PS I below.

Such coupling ensures that a total photocatalytic activity of the crystal comprises at least one fifth the sum of all individual photocatalytic activities. An exemplary photopotential for a layer composed of PS Is is approximately 1 volt.

According to one embodiment, the total photocatalytic activity of the crystal equals X times the sum of individual photocatalytic activities of all the layers in the crystal, where X is at least 0.2, more preferably at least 0.25, more preferably at least 0.3, more preferably at least 0.4 more preferably at least 0.5 more preferably at least 0.6 more preferably at least 0.7 more preferably at least 0.8 more preferably at least 0.9 and even more preferably the total photocatalytic activity of the crystal comprises the sum of all individual photocatalytic activities. It will be appreciated that electron carriers (such as metal ions and organic electron carriers) or semi-conducting materials (e.g. plastic polymers or lios such as silicon) may be incorporated into the crystal of the present invention to further increase the total photocatalytic activity.

Exemplary metal ions that may be incorporated into the crystal of the present invention include heavy metal ions such as platinum, and other metal ions such as silver, electron carriers such as methyl viologen and any other electron carrier.

Thus, according to another embodiment, the total photocatalytic activity comprises the number of layers times the sum of all individual photocatalytic activities.

According to one embodiment, the crystal comprises at least 100 layers of PS Is. According to another embodiment, the crystal comprises at least 200 layers of PS Is. According to another embodiment, the crystal comprises at least 400 layers of PS Is. According to another embodiment, the crystal comprises at least 600 layers of PS Is. According to another embodiment, the crystal comprises at least 800 layers of PS Is. According to another embodiment, the crystal comprises at least 1000 layers of PS Is. According to another embodiment, the crystal comprises more than 1000 layers of PS Is.

The crystal of the present embodiments may comprise a light energy conversion efficiency of about 20 %, more preferably 22 %, more preferably 24 %, more preferably 26 %, more preferably 28 %, more preferably 30 %, more preferably 32 %, more preferably 34 %, more preferably 36 %, more preferably 38 %, more preferably 40 %, more preferably 42 %, more preferably 44 %, and even more preferably 47 %.

According to an embodiment of the present invention, the crystal is characterized by a p21 space group. In various exemplary embodiments of the invention the β angle characterizing this space group is from about 90.8° to about 91.4°. For example, the unit cell can have the following dimensions: $a = 181.90 \pm 10 \text{ \AA}$, $b = 190.24 \pm 10 \text{ \AA}$ and $c = 219.66 \pm 10 \text{ \AA}$. A second arrangement in P21 space group the dimension of the space group are $a=124.9 \pm 5 \text{ \AA}$, $b=187.3 \pm 5 \text{ \AA}$, $c=132.0 \pm 5 \text{ \AA}$ $\beta=91 \pm 1^\circ$.

As used herein, the term "space group" refers to a group or array of operations consistent with an infinitely extended regularly repeating pattern. It is the symmetry of a three-dimensional structure, or the arrangement of symmetry elements of a crystal. There are 230 space group symmetries possible; however, there are only 65 space group symmetries available for biological structures. See, for example, U.S. Appl. No. 2004/0002145.

As used herein, the term "unit cell" refers to the fundamental portion of a crystal structure that is repeated infinitely by translation in three dimensions. A unit cell is characterized by three vectors **a**, **b**, and **c**, not located in one plane, which form the edges of a parallelepiped. The angles α , β and γ define the angles between the vectors: α is the angle between vectors **b** and **c**; β is the angle between vectors **a** and **c**; and γ is the angle between vectors **a** and **b**. The entire volume of a crystal can be constructed by regular assembly of unit cells. Each unit cell comprises a complete representation of the unit of pattern, the repetition of which builds up the crystal. See, for example, U.S. Appl. No. 2004/0002145.

According to still another embodiment the crystal comprises dimensions of about 500x100x10 μM .

The structure of the crystal may comprise coordinates of the protein databank files (PDB) files 1QZV (<http://www.dotsbdtorg/pdb/explore/exploredotdo?structureId=1QZV>) and 2001

(<http://www.dtrcsb.dootorg/pdb/explore/exploredotdo?structureId=2O01>), along with the unit cell dimensions provided herein.

According to other embodiments, the structure of the crystal remains intact following three minute incubation with a phosphate buffer (e.g. at pH 6-6.5) at room temperature.

According to other embodiments, the structure of the crystal remains intact following a five minute incubation with a phosphate buffer at room temperature.

According to other embodiments, the structure of the crystal remains intact following a ten minute incubation with a phosphate buffer at room temperature.

The present invention also contemplates a method of generating the above described crystals.

Thus, according to another aspect of the present invention there is provided a method of crystallizing pea *Pisum sativum* PS I the method comprising:

(a) suspending a purified preparation of the PS I in a crystallization solution, the crystallization solution comprising:

- (i) an aqueous buffer;
 - (ii) PEG 400;
 - (iii) a salt; and
 - (iv) PEG 6000 at a gradient of about 3.5-7 % w/v,
- under conditions suitable to generate crystallized PS I;

(b) contacting the crystallized PS I with a protecting solution, the protecting solution comprising PEG 400 and PEG 6000, thereby crystallizing the PSI.

As used herein, the phrase "purified preparation of the PS I" refers to a preparation of PS I that has been at least partially removed from its natural site of synthesis (e.g., whole pea). Preferably the PS I is substantially free from substances (e.g., other cells, proteins, nucleic acids, etc.) that are present in its *in-vivo* location. An exemplary method for growing pea plants such that isolation of undamaged thylakoids with unified PS I proteins are generated including methods of removing PS I from the pea plant is described in Example 1 of the Examples section hereinbelow. The present invention also envisages using any other methods of purification and isolation so long as the PS I remains functional. The PS Is may be fully purified or part

of a membrane preparation. Methods of preparing membrane extracts are well known in the art. For example, Qoronfle et al., [J Biomed Biotechnol. 2003; 2003(4): 249–255] teach a method for selective enrichment of membrane proteins by partition phase separation. Various kits are also commercially available for the preparation of
5 membrane extracts such as from Sigma-Aldrich (ProteoPrep™ Membrane Extraction Kit).

According to one embodiment the concentration of PS I in the crystallization solution is about 9 mg/ml.

According to yet another embodiment, the aqueous buffer comprises 20 mM
10 MES-bis-Tris pH 6.3, 0.5 % (v/v).

According to yet another embodiment, the crystallizing salt comprises ammonium succinate or ammonium citrate (e.g. at about 10 mM).

Preferably the protecting step is effected about 9 days following the initial crystallizing.

According to yet another embodiment, the protecting solution further comprises
15 a buffer (e.g. 20 mM MES-bis-Tris pH 6.6, 0.5 % (v/v) and a salt (e.g. ammonium succinate or ammonium citrate, for example at a concentration of about 10 mM).

As mentioned, the crystal of the present invention can also be generated with a conducting or semi-conducting material incorporated between the oriented layers of the
20 crystal.

According to this embodiment, the crystal may be soaked in a solution of metal ions during or following the crystallization process, preferably prior to the protecting stage. Semi-conducting material may be incorporated in aqueous solution or evaporation into the dry crystal.

Following crystal generation, the PS Is of the present invention may be attached
25 to a solid surface by covalent or non-covalent bonding (electrostatic). As used herein the term "covalent bond" refers to the linkage of two atoms by the sharing of two electrons, one contributed by each of the atoms. According to one embodiment, the crystal unit is bonded indirectly to a solid surface (i.e. using a biofunctional connecting
30 molecule).

It will be appreciated that in order to contact the PS I with a solid surface, it is preferable that the crystal does not contain PEG. Thus, prior to contacting, the present

invention envisages rinsing the crystal in a buffer which is devoid of PEG (e.g. in a phosphate buffer, such as a sodium or potassium phosphate buffer. The rinsing may be effected for 2 minutes, 3 minutes or even 5 minutes. According to one embodiment, the rinsing is not effected for more than 10 minutes.

5 The bifunctional connecting molecule may be attached to a free carboxyl of the PS I, a free primary amine of the PS I and/or to a thiol group in the PS I.

According to one embodiment the bifunctional connecting molecule comprises a succinylimide moiety.

The solid surface may be a conducting material, such as a transition metal. Examples of transition metals which may be used according to this aspect of the present invention include, but are not limited to silver, gold, copper, platinum, nickel, aluminum and palladium.

According to another embodiment, the solid surface is a semiconductor material (e.g. silicon).

A silicon surface may be modified by chemisorption of silan amine. The free amine groups may then be covalently bonded to the free carboxyls of the PS I on the surface of the crystals by carbodiimide chemistry. Reaction in aqueous solution pH 7, containing 1-Ethyl-3-[3-dimethylaminopropyl]carbodiimide hydrochloride (EDC or EDAC) connects the free amines on the surface of the silicon with the carboxyls of the PS-I protein when the crystal are laid on top of the modified semiconductor surface. Alternatively, the free amine groups of silane amine may be covalently bound to the free primary amines of the protein by a short connecting molecule Sulfo-MBS (m-Maleimidobenzoyl-N-hydroxysulfosuccinimide ester) in which the succinimide is first bound to the silanamine at the surface to form an oriented monolayer. Next, the maleinide end of the molecule binds (at pH 8) the free amines of the crystallized PS I. Such covalent binding of the PSI crystals to the surface of the gate form an active coupled electronic junction.

According to this aspect of the present invention, the crystallized PS Is retains photocatalytic activity following attachment to a solid surface.

Herein, the phrase "photocatalytic activity" refers to the conversion of light energy to electrical energy. Preferably, the crystallized PS Is retain at least 20 %, more preferably at least 30 %, more preferably at least 40 %, more preferably at least 50 %,

more preferably at least 60 %, more preferably at least 70 %, more preferably at least 80 %, more preferably at least 90 %, e.g., about 100 % the activity of a crystallized PS I prior to attachment to a solid surface. The present invention also envisages that the immobilized crystallized PS Is of the present invention comprises an activity greater
5 than that of non-immobilized PSIs.

Methods of measuring photocatalytic activity on surfaces fabricated therewith include measuring the photovoltage properties of the fabricated surfaces. The photovoltage properties may be measured for example by Kelvin probe force microscopy (KPFM). Photocatalytic activity may also be measured by analyzing the
10 electron transfer in the PS Is. Electron transfer may be measured by analyzing flash-induced absorption changes as measured by single turnover spectroscopy.

An experimental example of a photopotential generated by PS I crystals bound through connecting molecules to a gold electrode as described in FIG. 5A, is given in Figures 11A-B. The KPFM measurements show fast surface potential generation on
15 turning light on. The photopotential built up to 12 V whereby the instrument measurement capacity reached saturation. Thus, the generated photopotential was much larger than 12 V, but could not be measured by the KPFM instrument used. On turning the light off there was a reduction of the photopotential. The photopotential was generated with opposite polarity depending whether the PS I crystals were bound to the
20 electrode through the oxidizing end (Figure 11A) or through the reducing end (Figure 11B) of the crystal.

FIG. 12A is a graph showing the experimental measurements of a photopotential generated by hybrid PS I crystals bound to n-GaAs surface. An electronic junction between the PS I crystals and the semiconductor were formed by a self-assembled
25 monolayer of linker molecules. The light induced photovoltage decreases the band bending and allows current flow between drain and source (FIG. 3G). The unprecedented very high voltage generated by the PS I-crystals/semiconductor junction paves the way to fabricate extremely sensitive photon counting light sensors.

Reference is now made to FIG. 6 which is a schematic illustration of an
30 optoelectronic device **10**, according to various exemplary embodiments of the present invention. Device **10** comprises one or more photocatalytic active crystals **12** interposed between a first electrode **14** and a second electrode **16**. In various exemplary

embodiments of the invention at least one of the photocatalytic active crystal(s) **12** comprises serially oriented layers of pea Pisum sativum Photosystem I (PS I), wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all individual photocatalytic activities, as further detailed hereinabove. Photocatalytic active crystal(s) **12** can be bonded to electrodes **14** and/or **16**, directly or indirectly, by covalent or non-covalent bonding, as further detailed hereinabove. The bonding between crystal **12** and electrode **14** and **16** is preferably such that electronic junctions **18** are formed on both sides of crystal **12**.

In various exemplary embodiments of the invention the work functions of electrodes **14** and **16** differ. Generally, electrode **14** injects electrons to crystal **12** serves as a source of electrons and electrode **16** serves as a source of holes. Suitable materials for electrode **14** include, without limitation, metals such as Au, Ag, Ni, Ti and Al or any other metal having similar properties. Suitable materials for electrode **16** include, without limitation, materials having any one of InSnO_2 , SnO_2 and zinc oxide (ZnO) metal alloys. Other than these alloys, oxides of Sn and Zn may also be contained in the material of electrode **16**. In some embodiments of the present invention electrode **16** is made of Indium tin oxide (ITO).

At least one of electrodes **14** and **16** is preferably transparent to light at wavelength or range of wavelengths which induce charge separation within crystal **12**. In an embodiment of the present invention, electrode **16** is transparent and electrode **14** is opaque. For example, electrode can be made of gold and electrode **16** can be made of TIO. Device **10** can also comprise a substrate **20** on which electrode **14** can be deposited. In an embodiment of the invention substrate **20** is made of glass, but other materials such as, but not limited to, SiO_2 , plastics such as polyphthalamide, polyarylamide, polysulfone, polyethersulfone, polyphenylsulfone, modified polyphenylsulfone, polyamide-imide, can also be used. In some embodiments of the present invention device **10** also comprises a cover substrate **22** on top of electrode **16**. Substrate **22** is preferably transparent to light at wavelength or range of wavelengths which induce charge separation within crystal **12**. Substrate **22** can serve for protecting electrode **16** from the environment and can be made of any transparent material such as glass, transparent polymer, etc. In some embodiments of the present

invention device **10** comprises a protective layer **26** interposed between crystal **12** and electrode **16**. Protective layer **26** is preferably transparent to light at wavelength or range of wavelengths which induce charge separation within crystal **12**. Layer **26** can be made of a conductive n-type polymer which can be deposited on crystal **12**, *e.g.*, by spin coating technique, before the layering of the electrode **16**.

Device **10** can be shaped as a cavity having a base and walls formed in an isolating substrate, wherein first electrode **14** is deposited onto the base, and second electrode **16** is supported by the walls. In the representative example of FIG. 6, the cavity is defined between substrate **22** and walls **24** which can be made of silicon nitride or any other dielectric material. For example, a cavity can be formed in a dielectric layer by photolithography followed by etching. The dielectric layer can be deposited on the first electrode and the etching can be performed such as to expose the first electrode on the base of the cavity.

Device **10** can be used in the field of micro- and sub-microelectronic circuitry and devices including, but not limited to, spatial imaging devices, solar batteries, optical computing and logic gates, optoelectronic switches, diodes, photonic A/D converters, and thin film "flexible" photovoltaic structures.

For example, in an embodiment of the invention, device **10** serves as a photodiode. In the absence of activation light (*e.g.*, in the dark) crystal **12** serves as a diode. Once stimulated by light at the appropriate wavelength or range of wavelength, charge separation occurs within crystal **12** and device **10** functions as a photodiode. The operation of device **10** according to this embodiment is illustrated in FIG. 4, showing a dark current (J_{dark}) and a photocurrent (J_{sc}) flowing through a shunt resistor (R_{sh}) and a series resistor (R_{s}).

In use, crystal **12** is irradiated by light hence being excited to efficient charge separation of high quantum efficiency (*e.g.*, above 95 %). Electrodes **14** and **16** tap off the electrical current caused by the charge separation. Depending on the voltage applied between electrodes **14** and **16**, device **10** can be used either as a photovoltaic device, or as a reversed bias photodiode.

Specifically, in the absence of external voltage, device **10** enacts a photovoltaic device which produces current when irradiated by light. Such device can serve as a component in, *e.g.*, a solar cell.

When reverse bias is applied between electrodes **14** and **16**, device **10** maintains high resistance to electric current flowing from contact **14** to contact **16** as long as device **10** is not irradiated by light which excites crystal **12**. Upon irradiation by light at the appropriate wavelength or range of wavelengths, the resistance is significantly reduced. Such device can serve as a component in, *e.g.*, a light detector.

Optoelectronic device **10** can also serve as a solar cell, when no bias voltage is applied. Upon irradiation of crystal **12**, the charge-separated state results in internal voltage between donor site **16** and acceptor site **18**. The internal voltage can be tapped off via electrical contacts at donor site **16** and acceptor site **18**. If the current circuit is closed externally, the current flow is maintained through repeated light-driven charge separation in the solar cell.

FIG. 7 is a schematic illustration of a photogate or light sensor device **40**, according to various exemplary embodiments of the present invention. Device **40** comprises a semiconductor electrode surface **42** having thereon one or more photocatalytic active crystals **12**, as further detailed hereinabove. In some embodiments of the invention crystal(s) **12** are bound to surface **42** by non-direct covalent binding so as to form an electronically coupled junction. Light absorbed by the crystals induces a potential on surface **42** thereby allowing photogating at high frequency.

In some embodiments of the present invention a device comprising crystal(s) **12** (*e.g.*, device **10** or device **40**) serves as an optocoupler. This embodiment is particularly useful for transferring signals from one element to another without establishing a direct electrical contact between the elements, *e.g.*, due to voltage level mismatch. For example, a device comprising crystal(s) **12** can be used to establish contact free communication between a microprocessor operating at low voltage level and a gated switching device operating at high voltage level.

Thus, for example, one element can include an optical transmitter (not shown), such as, but not limited to, a light emitting diode (LED), while another element can include or be electrically coupled to a device comprising crystal(s) **12** (*e.g.*, device **10** or **40**). Crystal **12** is selected such that the radiation emitted by the optical transmitter induces charge separation between in crystal **12**. In use, the transmitter and crystal **12** are kept at optical communication but are electrically decoupled. Light emitted by the

transmitter is received by a device comprising crystal **12** and is converted to electrical signal by means of charge separation, as further detailed hereinabove.

FIGs. 8A-B are schematic illustrations of a field effect transistor (FET) device **50**, which according to some embodiments of the present invention, can be photogated
5 by photocatalytic active crystal **12**.

FET device **50** comprises a substrate **60** formed with a source region **52** and a drain region **54**. Regions **52** and **54** are laterally displaced. The polarity of substrate **60** is opposite to the polarity of regions **52** and **54**. Specifically, when substrate **60** has a p-type polarity, each of regions **52** and **54** has an n-type polarity, and when substrate **60**
10 has an n-type polarity, each of regions **52** and **54** has a p-type polarity. Crystal(s) **12** are deposited on the surface **66** of substrate **60**, at least between regions **52** and **54**, such that an electric junction is at surface **66** between the source and the drain. In some embodiments, crystal **12** extends further such that there is an overlap between crystal **12** and region **52** and/or **54**.

15 In some embodiments of the present invention, at least part of surface **66** is coated by an insulating layer **58** and crystals **12** are attached to layer **58**, *e.g.*, by covalent bond, as further detailed hereinabove. Layer **58** preferably coats surface **66** between regions **52** and **54** but may also extend further such that there is an overlap between layer **58** and region **52** and/or **54**.

20 FET device **50** may further comprise electrodes **62** and **64** for contacting regions **52** and **54**, respectively. The electrodes can be made of electrically conductive material, such as a metal.

FIG. 8B illustrates the gating principle of device **50**. Gating of current flow between source **52** and drain **54** is achieved by irradiating crystal **12** by light. Light
25 induced charge separation in crystal **12** generates voltage at surface **66** resulting in injection of charge carriers from crystal **12** to substrate **60** and formation of a conduction channel **56** between source region **52** and drain region **54**. The layers of crystal **12** are constituted such that the charge carriers that are injected from crystal **12** to substrate **60** are of the same polarity as the source and the drain. Specifically, when
30 the source and drain are n-type regions, crystal **12** injects electrons to substrate **60** (hence forms an n-channel or depletion layer at **56**), and when the source and drain are

p-type regions, crystal 12 injects holes to substrate 60 (hence form a p-channel at 56).

In embodiments in which insulating layer 58 is employed, light induced charge separation in crystal 12 generate electric dipole in substrate 60 (near surface 66) which generates a depletion layer that serves as channel 56, similarly to the voltage gate in conventional MOSFET. In these embodiments, the layers of crystal 12 are constituted such that the depletion layer is of the same polarity as the source and the drain. Specifically, when the source and drain are n-type regions crystal 12 forms an n-type depletion layer at 56, and when the source and drain are p-type regions crystal 12 forms a p-type depletion layer at 56).

FIG. 3G is a representative example of a band diagram, which, corresponds, without limitation, to an n-channel FET. One of ordinary skill in the art would know how to construct a diagram for the case of a p-channel FET. For clarity of presentation, also provided is FIGs. 3B-C which are a schematic illustration (FIG. 3B) and a band diagram of a traditional MOSFET. Referring first to FIG. 3C, at zero gate bias (black lines), the semiconductor channel is partially depleted. When a more positive gate bias is applied (red lines), more charges are induced in the channel via capacitive coupling thus resulting in a larger current flowing between the source and drain. Referring now to FIG. 3G, the third terminal of the FET device of the present embodiments, as stated, comprises crystal 12. Upon absorbing photons, electrons transfer into the semiconductor thus increasing the conductivity of the channel. Holes can also migrate to the crystal 12 so as to attract more electrons supplied from the ohmic contacts. Therefore, an open channel results (red lines in FIG. 3G). The induced photocurrent in the FET device of the present embodiments is typically proportional to the ratio of the hole lifetime over the electron transit time from the source to the drain, and its switching characteristics depends on the two time constants.

FET device 50 can serve as an amplifier or a switching device wherein the light controls the current flowing from the source region to the drain region. A symbolic illustration of the FET device 50 is illustrated at the right hand side of FIG. 2.

Representative examples of semiconductor materials suitable for device **50**, include, without limitation, Si, SiC, SiGe, GaAs, AlGaAs, InGaAs, InGaP, AlInP, GaN and Ge.

As will be appreciated by one ordinarily skilled in the art, FET device **50** can
5 operate without gating voltage (namely, while the crystal **12** is left as an open terminal) because the gating is induced by photons impinging on crystal **12**. Device **50** can be used as a logical element whereby the FET can be switched to an "on" state by the incident light. In addition, device **50** can be used as the backbone of an image sensor with large patterning possible due to a strong variation of the drain current with the
10 spatial position of the incident light beam. Several phototransistors like device **50**, each operating at a different wavelength can be assembled to allow sensitivity of the image sensor to color images. The charge storage capability of the structure with further modifications known to one skilled in the art of conventional semiconductors can be exploited for memory related applications.

15 Devices **10**, **40** and/or **50** can be integrated in many electronic circuitries. In particular, such devices can be used as building blocks which can be assembled on a surface structure to form a composite electronic assembly. For example, two or more photodiodes or phototransistors can be assembled on a surface structure to form a logic gate, a combination of logic gates or a microprocessor.

20 FIG. 9 is a flowchart diagram of a method suitable for fabricating an optoelectronic device, according to various exemplary embodiments of the present invention. It is to be understood that, unless otherwise defined, the operations described hereinbelow can be executed either contemporaneously or sequentially in many combinations or orders of execution. Additionally, one or more operations described
25 below are optional and may not be executed.

The method begins at **90** and optionally and preferably continues to **91** at which the method forms a cavity having a first electrode deposited on the base of the cavity. In some embodiments of the present invention the method proceeds to **92** at which the first electrode is passivated so as to allow binding of one or more photocatalytic active
30 crystals thereon. The passivation can be for example, by attaching bifunctional connecting molecules on a surface of the first electrode. Optionally and preferably the

bifunctional connecting molecules form a sulfide bond with first electrode and an amide bond with crystal.

The method continues to **93** at which one or more photocatalytic active crystals are deposited on a first electrode. In various exemplary embodiments of the invention at least one of the photocatalytic active crystal(s) comprises serially oriented layers of pea Pisum sativum Photosystem I (PS I), wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all individual photocatalytic activities, as further detailed hereinabove.

In some embodiments of the present invention the method continues to **94** at which the crystal is coated by a protective coat or layer. The protective coat or layer is preferably transparent to light at wavelength or range of wavelengths which induce charge separation within the crystal. The protective coat or layer can be made of a conductive n-type polymer. The coating can be by means of spin coating.

In various exemplary embodiments of the invention the method continues to **95** at which a second electrode is deposited on the crystal. In embodiments in which a cavity is formed, the deposition is preferably such that the second electrode is supported by the walls of the cavity.

The method ends at **96**.

FIG. 10 is a flowchart diagram of a method suitable for fabricating a transistor, according to various exemplary embodiments of the present invention. It is to be understood that, unless otherwise defined, the operations described hereinbelow can be executed either contemporaneously or sequentially in many combinations or orders of execution. Additionally, one or more operations described below are optional and may not be executed.

The method begins at **100** and optionally and preferably continues to **101** at which a surface of a substrate is modifying so as to allow binding of one or more photocatalytic active crystals thereto. The substrate is preferably formed with a source region and a drain region as further detailed hereinabove.

In some embodiments of the present invention the modification comprise chemisorption of surface modifying molecules which comprise free amine groups. The surface modifying molecules are preferably selected to bind directly to the crystal. A

representative example of suitable surface modifying molecules includes, without limitation, silan amine. In some embodiments of the present invention the method continues to **102** at which the modified surface is contacted with connecting molecules such as to form an amide bond between the surface modifying molecules and the connecting molecules. A representative example of suitable connecting molecules includes, without limitation, Sulfo-MBS.

Alternatively to **101** and **102**, the method can continue to **103** at which an isolating layer is formed on the surface so as to allow binding of the crystal to the isolating layer. A representative example of suitable isolating layer includes, without limitation, a silicon oxide, hafnium silicate, zirconium silicate, hafnium dioxide and zirconium dioxide layers.

The method continues to **104** at which one or more photocatalytic active crystals are electrically coupled to the substrate at least between the source region and the drain region. In various exemplary embodiments of the invention at least one of the photocatalytic active crystal(s) comprises serially oriented layers of pea Pisum sativum Photosystem I (PS I), wherein each of the oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all individual photocatalytic activities, as further detailed hereinabove.

The coupling is preferably so as to bind the crystal to then surface either directly or through a binding medium which can be surface modifying molecules, connecting molecules or isolating layer.

The method ends at **105**.

Additional objects, advantages and novel features of the present invention will become apparent to one ordinarily skilled in the art upon examination of the following examples, which are not intended to be limiting. Additionally, each of the various embodiments and aspects of the present invention as delineated hereinabove and as claimed in the claims section below finds experimental support in the following examples.

EXAMPLES

Reference is now made to the following examples, which together with the above descriptions, illustrate the invention in a non limiting fashion.

5 Generally, the nomenclature used herein and the laboratory procedures utilized in the present invention include molecular, biochemical, microbiological and recombinant DNA techniques. Such techniques are thoroughly explained in the literature. See, for example, "Molecular Cloning: A laboratory Manual" Sambrook et al., (1989); "Current Protocols in Molecular Biology" Volumes I-III Ausubel, R. M., ed. 10 (1994); Ausubel et al., "Current Protocols in Molecular Biology", John Wiley and Sons, Baltimore, Maryland (1989); Perbal, "A Practical Guide to Molecular Cloning", John Wiley & Sons, New York (1988); Watson et al., "Recombinant DNA", Scientific American Books, New York; Birren et al. (eds) "Genome Analysis: A Laboratory Manual Series", Vols. 1-4, Cold Spring Harbor Laboratory Press, New York (1998); 15 methodologies as set forth in U.S. Pat. Nos. 4,666,828; 4,683,202; 4,801,531; 5,192,659 and 5,272,057; "Cell Biology: A Laboratory Handbook", Volumes I-III Cellis, J. E., ed. (1994); "Culture of Animal Cells - A Manual of Basic Technique" by Freshney, Wiley-Liss, N. Y. (1994), Third Edition; "Current Protocols in Immunology" Volumes I-III Coligan J. E., ed. (1994); Stites et al. (eds), "Basic and Clinical Immunology" (8th 20 Edition), Appleton & Lange, Norwalk, CT (1994); Mishell and Shiigi (eds), "Selected Methods in Cellular Immunology", W. H. Freeman and Co., New York (1980); available immunoassays are extensively described in the patent and scientific literature, see, for example, U.S. Pat. Nos. 3,791,932; 3,839,153; 3,850,752; 3,850,578; 3,853,987; 3,867,517; 3,879,262; 3,901,654; 3,935,074; 3,984,533; 3,996,345; 4,034,074; 25 4,098,876; 4,879,219; 5,011,771 and 5,281,521; "Oligonucleotide Synthesis" Gait, M. J., ed. (1984); "Nucleic Acid Hybridization" Hames, B. D., and Higgins S. J., eds. (1985); "Transcription and Translation" Hames, B. D., and Higgins S. J., eds. (1984); "Animal Cell Culture" Freshney, R. I., ed. (1986); "Immobilized Cells and Enzymes" IRL Press, (1986); "A Practical Guide to Molecular Cloning" Perbal, B., (1984) and 30 "Methods in Enzymology" Vol. 1-317, Academic Press; "PCR Protocols: A Guide To Methods And Applications", Academic Press, San Diego, CA (1990); Marshak et al., "Strategies for Protein Purification and Characterization - A Laboratory Course

Manual" CSHL Press (1996); all of which are incorporated by reference as if fully set forth herein. Other general references are provided throughout this document. The procedures therein are believed to be well known in the art and are provided for the convenience of the reader. All the information contained therein is incorporated herein
5 by reference.

EXAMPLE 1

Preparation of serially oriented plant PS I crystals

10 MATERIALS AND METHODS

The preparation of the oriented crystals consisted of three stages, which included plant growth, isolation of PS I and crystallization thereof.

Plant growth and isolation of thylakoids from leaves: The first stages of the technology were designed to develop plant growth conditions that would optimize the
15 isolation of undamaged thylakoids with unified PS I proteins from which intact and unified plant PS I could be isolated. Peas (*Pisum sativum*, var. Alaska) grew for 12 days under cool-white fluorescent light (90-130 μ En) in a 16 hours light/ 8 hours dark cycle. All subsequent steps were carried out at 4 °C. Washed leaves (60-75 gr) were ground with ice-cold STN buffer (0.3 M sucrose, 15 mM NaCl, 30 mM Tricine-NaOH
20 pH 8, 1 mM PMSF, 15 μ M Leupeptin, 1 μ M pepstatin A) for 15 seconds. The slurry was filtered through 6 layers of cheesecloth and centrifuged for a few seconds. Chloroplasts were then precipitated by centrifugation at 2500 x g for 7 min and suspended in a hypotonic medium (5 mM Tricine-NaOH pH 8). Thylakoids were collected by centrifugation at 20,000 x g for 10 min and re-suspended in a buffer containing 5mM
25 Tricine-NaOH pH 8 and 150 mM NaCl. Thylakoid membranes were then precipitated and re-suspended in minimal volume of STN2 buffer (0.3 M sucrose, 20 mM Tricine-NaOH pH 8, 1mM PMSF). The thylakoid concentration was adjusted to 3 mg chlorophyll /ml and the detergent Dodecyl-maltoside (Anatrace) was added to a final concentration of 0.5 % w/v. After 10 minutes of incubation, detergent treated thylakoid
30 membranes were collected by ultra-centrifugation at 150,000 xg for 30 minutes, the pellet was re-suspended in minimal volume STN2 buffer and stored

at -80°C . PS I was extracted from thylakoid by the detergent Dodecyl-maltoside and collected by ultra-centrifugation (10).

Isolation of PS I from thylakoids: PS I was isolated as previously described (10) or by alternative novel method as follows: A negative purification to remove the b6f complex, and F-ATPase was performed by incubating thylakoids at 2.6 mg chlorophyll per ml with 0.5 % DM followed by centrifugation at 150,000 g for 30 minutes. The resulting pellet was immediately suspended by glass-Teflon homogenizer in a buffer containing 0.3 M sucrose, 20 mM Tricine (pH 8) and 1 mM PMSF) at chlorophyll concentration of about 3 mg/ml. PS I was solubilized by the addition of 10 % dodecyl maltoside (DM) to give about 1.8 % (6.0 mg detergent/mg chlorophyll) final concentration. Unsolubilized material was removed by centrifugation at 150,000 g for 15 minutes and all the following steps were performed at 0°C to 4°C in darkness or dim light. The supernatant was applied on a DEAE-cellulose (Whatman DE52) column of 1.5 cm diameter and length of approximately 0.5 cm/mg chlorophyll that was washed with 30 ml buffer containing 20 mM Tricine-Tris (pH 7.4) and 0.2 % DM. The loaded column is washed with the same buffer and PS I was eluted (flow rate of $>2\text{ml/min}$) by a linear gradient of 0 to 300 mM NaCl (100 ml in each chamber) in the same buffer. Fractions of 5 ml were collected and usually four of them represent about half of the eluted dark-green peak were combined and PS I was precipitated by the addition of 50% PEG 6000 (Hampton) to give final concentration of 10%. The pellet obtained by centrifugation at 10,000 g for 10 min was solubilized by 4 ml buffer containing 20 mM Tricine-Tris (pH 7.4) and 0.05% decylthiomaltoside for further purification sucrose gradient centrifugation as described (10) except that decylthiomaltoside substituted for dodecylthiomaltoside.

Crystallization of purified PS I: Crystallization was performed using the sitting-drop variant of the vapour-diffusion technique at 4°C . $4\mu\text{l}$ drops of purified protein at 2.5mg chlorophyll / ml ($\sim 9\text{mg protein/ml}$) were mixed with equal volumes of the 0.5 ml reservoir solution [20 mM MES-bis-Tris pH 6.3, 0.5% (v/v) PEG 400, 10 mM ammonium succinate, and a gradient of 3.5 to 7 % (w/v) PEG 6000 and 0.05 % decylthiomaltoside]. Crystals appeared within two-three days and after nine days were treated by a cryo-protecting solution containing 20 mM MES-bis-Tris pH 6.6, 0.5% (v/v) PEG 400, 10 mM ammonium citrate, and gradually increased 10 to 20%

(w/v) PEG 6000 in steps of 10, 15 and 20 %. The crystals in space group P21, with unit-cell parameters $a=124.9 \pm 5 \text{ \AA}$, $b=187.3 \pm 5 \text{ \AA}$, $c=132.0 \pm 5 \text{ \AA}$ $\beta=91 \pm 1^\circ$, and crystal lattice are stable for several months at 10 °C. This unique procedure yielded highly ordered crystals in which PS I molecules were serially oriented in 1000 layers in a crystal.

EXAMPLE 2

Generation of a photogate using the crystals of the present invention

Electronically active junctions between plant PS I crystals and a semiconductor surface can be used to fabricate a sensitive light sensor. In this embodiment, the present inventors propose that the crystals may be bound to a semiconductor surface by non-direct covalent binding thereby forming an electronically coupled junction. This may be effected for example, through a chemisorbed monolayer of small connecting molecules on the semiconductors surfaces and nanotubes, in a similar fashion to non-crystallized PS I binding to a surface (See Figures 1B-C). The photoactive crystals can be used to replace the conventional metallic gates (Figure 2). In such devices the application of the gate voltage induces a conducting channel and enabling current flow from drain to source (Figures 2 and 3B-C). The current flow is exponential with the applied gate voltage which is the basis for amplification (Figure 3A).

EXAMPLE 3

Fabrication of micro photovoltaic cells and microphoto-switches using the crystals of the present invention

Photovoltaic devices may be fabricated by the formation of electronic junctions on both sides of the oriented multilayers of PS I in the microcrystals. PS I microcrystals behave like a diode in the dark and photodiode in the light, and therefore can be used when placed between two electrodes as a photovoltaic cell. They functions as schematically presented in Figure 4. Such a device can be described as consisting a diode, dark current (R_{dark}), photocurrent (J_{sc}) going through a shunt (R_{sh}) and series resistor (R_{s}). A detailed schematic example for fabrication of PS I crystal based photovoltaic cell is represented in Figures 5A-B. The device consists of a bottom 200

nm metal electrode made of Au, Ag, Ni, Ti, Al or similar metals deposited on flat solid substrate such as glass, silicon oxide, plastic or similar materials. The metal electrode is passivated by a small bifunctional connecting molecule through the formation of a sulfide bond between a thiol at the one end of the molecule. The other end consists of an amine-binding group such as succinylimide or similar primary amine reactive group. A second electrode is deposited on the crystal of photoactive PS I. The second electrode is preferably a hole-injection light transmissive electrode and it can be any electrode as long as it is capable of functioning as an anode to inject holes into the crystal. Preferably, the second electrode comprises InSnO_2 , which can be deposited by sputtering, electron beam vapor deposition, ion plating, indirect evaporation process. When required, a protective transparent conductive n-type polymer layer may be spun coated on the crystals before the layering of the top transparent electrode.

EXAMPLE 4

Fabrication of photo gate on a FET transistor

The assembly is fabricated by formation of a photogate made out of serially oriented multi-layers of PS I in crystals on surfaces of n channel of FET transistor. Light absorbed by the PS I crystals induces photo potential of 1 V / PS I. Current flow between source and drain in transistors such as PNP, NPN, PIN and similar can be switched on and off at MHz frequency by light absorbed by PS I attached to the base section. Devices fabricated by this bio-solid state hybrid can be used as photo gates and photo sensors in logic systems, optical communications and detection systems.

Below, is a detailed example describing the fabrication of Si FETs with the crystals of the present invention. The photogate on top of the n channel of an $\text{N}^{++}/\text{N}/\text{N}^{++}$ FET is made of PS I crystals (Figures 3D-F). The n+-channel is formed as a result of gating in the n-doped matrix. An electronic junction between the PS I crystals and the semiconductor surface is fabricated by a self-assembled monolayer of linker molecules. The silicon surface is modified by chemisorption of silan amine. The free amine groups are covalently bound to the free carboxyls of the PS I on the surface of the crystals by carbodiimide chemistry. Reaction in an aqueous solution pH 7, containing 1-Ethyl-3-[3-dimethylaminopropyl]carbodiimide hydrochloride (EDC or EDAC) connects the free amines on the surface of the silicon with the carboxyls of

the PS I protein when the crystal are laid on top of the modified semiconductor surface. Alternatively the free amine groups of silane amine are covalently bound to the free primary amines of the protein by a short connecting molecule Sulfo-MBS (m-Maleimidobenzoil-N-hydroxysulfosuccinimide ester) in which the succinimide is first bound to the silanamine at the surface of the transistor to form an oriented monolayer. Next, the maleinide end of the molecule binds (at pH 8) the free amines of the protein crystal. Such covalent binding of the PS I crystals to the surface of the gate form an active coupled electronic junction. Light induced charge separation in the PS I crystals generates a large photovoltage. This voltage causes the injection of negative charges from the crystal to the n channel lowering the band banding and opening the gate (Figure 3G) as previously demonstrated in a GaAs junction (4). Alternatively, the n channel is coated by insulating a silicon oxide layer that is covalently coupled to the PS I crystals. The photovoltage generated across 10 μm of dielectric crystal forms a large dipole positive at the surface of the n channel that opens the gate similar to voltage gate in MOSFET (Figure 3B). The properties of photogate are determined using Desert-cryogenics probe-station attached to a Keithley low current source measure unit equipped with a variable light source. The measurements are used to determine the efficiency of the electronic junction between the PS I crystals and the FET channel. They include light-induced amplification of current flow between drain and source and the quantum efficiency of the photogate.

EXAMPLE 5

Comparison between the crystals of the present invention and other PS I crystals

The photopotential of the crystals of the present invention were compared with the crystals generated as described in Amunts et al., Nature 2007, 447;58-63. The crystals of the present invention were grown the same way as described in Example 1, herein above except that the medium in which the crystals were grown comprised 0.015 % decyl maltoside. The PS I crystals were bound to a gold plated glass surface prior to measurement.

The results are portrayed in Figures 13A-B.

It is appreciated that certain features of the invention, which are, for clarity, described in the context of separate embodiments, may also be provided in combination in a single embodiment. Conversely, various features of the invention, which are, for brevity, described in the context of a single embodiment, may also be provided
5 separately or in any suitable subcombination.

Although the invention has been described in conjunction with specific embodiments thereof, it is evident that many alternatives, modifications and variations will be apparent to those skilled in the art. Accordingly, it is intended to embrace all such alternatives, modifications and variations that fall within the spirit and broad scope
10 of the appended claims. All publications, patents and patent applications mentioned in this specification are herein incorporated in their entirety by reference into the specification, to the same extent as if each individual publication, patent or patent application was specifically and individually indicated to be incorporated herein by reference. In addition, citation or identification of any reference in this application shall
15 not be construed as an admission that such reference is available as prior art to the present invention.

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WHAT IS CLAIMED IS:

1. A crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I (PS I), wherein each of said oriented layers comprises an individual photocatalytic activity and wherein a total photocatalytic activity of the crystal comprises at least the sum of all said individual photocatalytic activities.
2. The crystal of claim 1, further comprising a semi-conducting material in between said oriented layers.
3. The crystal of claim 1, wherein said semi-conducting material comprises a plastic polymer.
4. The crystal of claim 1, further comprising at least one electron carrier in between said oriented layers.
5. The crystal of claim 4, wherein said electron carrier is a metal.
6. The crystal of claim 1, comprising a β angle of about 90.8 to 91.4°.
7. The crystal of claim 1, characterized by having a space group of p21.
8. The crystal of claim 1, characterized by unit cell dimensions of $a = 181.90 \pm 10 \text{ \AA}$, $b = 190.24 \pm 10 \text{ \AA}$ and $c = 219.66 \pm 10 \text{ \AA}$.
9. The crystal of claim 1, wherein each of said layers comprises a photopotential of about 1V.
10. The crystal of claim 1, capable of light energy conversion efficiency of approximately 47 %.

11. The crystal of claim 1, wherein a surface of the crystal is attached to a bifunctional connecting molecule.

12. The crystal of claim 11, wherein said surface of said crystal is covalently attached to said bifunctional connecting molecule.

13. The crystal of claim 11, wherein said bifunctional connecting molecule is attached to a free carboxyl of said PS I.

14. The crystal of claim 11, wherein said bifunctional connecting molecule is attached to a free primary amine of said PS I.

15. The crystal of claim 11, wherein said bifunctional connecting molecule comprises a succinylimide moiety.

16. The crystal of claim 1, capable of withstanding contact with a phosphate buffer for at least 10 minutes without dissolving.

17. A method of crystallizing pea *Pisum sativum* PS I the method comprising:

(a) suspending a purified preparation of the PS I in a crystallization solution, said crystallization solution comprising:

- (i) an aqueous buffer;
- (ii) PEG 400;
- (iii) a salt; and
- (iv) PEG 6000 at a gradient of about 3.5-7 % w/v,
under conditions suitable to generate crystallized PS I;

(b) contacting said crystallized PS I with a protecting solution, said protecting solution comprising PEG 400 and PEG 6000,
thereby crystallizing the PS I.

18. The method of claim 17, wherein a concentration of said PS I in said crystallization solution is about 9 mg/ml.
19. The method of claim 17, wherein said salt comprises an ammonium salt.
20. The method of claim 17, wherein said protecting solution further comprises a buffer and a salt.
21. The method of claim 20, wherein said salt comprises an ammonium succinate salt.
22. The method of claim 17, wherein said protecting solution further comprises a semiconducting material or a conducting material.
23. A crystal comprising serially oriented layers of pea *Pisum sativum* Photosystem I (PS I), generated according to the method of claim 17.
24. A composition, comprising a solid surface attached to a plurality of crystals of claim 1.
25. The composition of claim 24, wherein said crystallized PS Is are directly attached to said solid surface.
26. The composition of claim 25 wherein said directly attached is covalently attached.
27. An optoelectronic device comprising at least one photocatalytic active crystal interposed between a first electrode and a second electrode, wherein said at least one photocatalytic active crystal is the crystal of claim 1.

28. An optoelectronic device comprising a plurality of photocatalytic active crystals interposed between a first electrode and a second electrode, wherein at least one of said photocatalytic active crystals is the crystal of claim 1.

29. A circuitry, comprising an arrangement of optoelectronic devices, wherein at least one of said optoelectronic devices is the optoelectronic device of claim 27 or 28 and wherein at least two of said optoelectronic devices are in electrical communication.

30. The device or circuitry of claim 27 or 28, serving as a component in a photodiode, a phototransistor, a photogate, a logic gate, a solar cell or an optocoupler.

31. The device or circuitry of any of claims 27-30, wherein at least one of said first electrode and said second electrode is transparent to light capable of activating said Photosystem.

32. The device or circuitry of any of claims 27-31, further comprising a cavity having a base and walls formed in an isolating substrate, wherein said first electrode is deposited onto said base, and said second electrode is supported by said walls.

33. A transistor, comprising a substrate formed with a source region and a drain region being laterally displaced from said source region, and a photogate attached to a surface of said substrate at least between said regions, wherein said photogate comprises the crystal of claim 1.

34. A method of fabricating an optoelectronic device, comprising depositing the crystal of claim 1 on a first electrode and depositing a second electrode on said crystal.

35. The method of claim 34, further comprising passivating said first electrode prior to said deposition of said crystal, so as to allow binding of said crystal to said first electrode.

36. The method of claim 35, wherein said passivation of said first electrode comprises attaching bifunctional connecting molecules on a surface of said first electrode.

37. The method of claim 36, wherein said bifunctional connecting molecules form a sulfide bond with said first electrode and an amide bond with said crystal.

38. The method of claim 34, further comprising coating said crystal with a protective coat prior to said deposition of said second electrode.

39. The method of claim 34, further comprising forming a cavity having a base and walls in an isolating substrate and depositing said first electrode onto said base, wherein said deposition of said second electrode is done such that said second electrode is supported by said walls.

40. A method of forming a transistor, comprising electrically coupling the crystal of claim 1 to at least an exposed surface of a substrate between a source region formed in said substrate and a drain region formed in said substrate, such that photo induced charge separation in said crystal results in formation of a conducting channel region between said source region and said drain region, thereby forming the transistor.

41. The method of claim 40, further comprising modifying said surface prior to said deposition of said crystal, so as to allow binding of said crystal to said surface.

42. The method of claim 41, wherein said modifying comprises chemisorption of surface modifying molecules which comprise free amine groups.

43. The method of claim 42, wherein said surface modifying molecules bind directly to said crystal.

44. The method of claim 42, further comprising contacting said modified surface with connecting molecules such as to form an amide bond between said surface modifying molecules and said connecting molecules.

45. The method of claim 44, further comprising generating conditions for binding of said connecting molecules to said crystal.

46. The method of claim 40, further comprising forming an isolating layer on said surface prior to said deposition of said crystal, so as to allow binding of said crystal to said isolating layer.

FIG. 1C

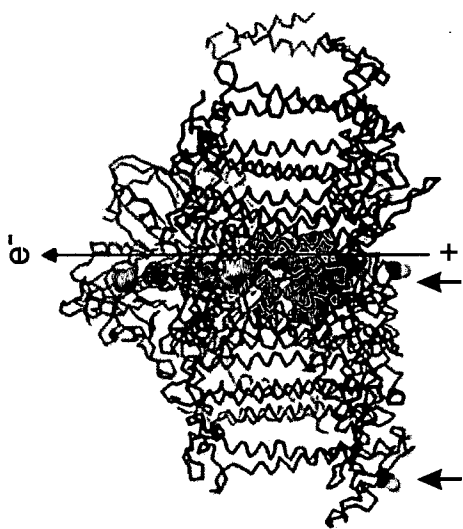
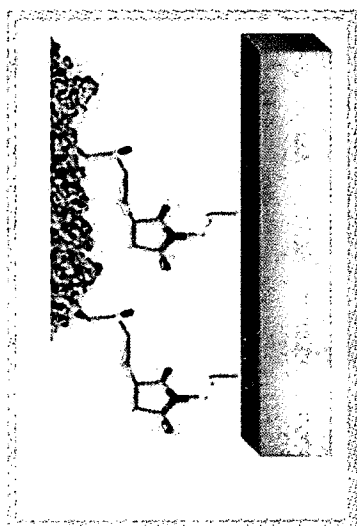


FIG. 1A

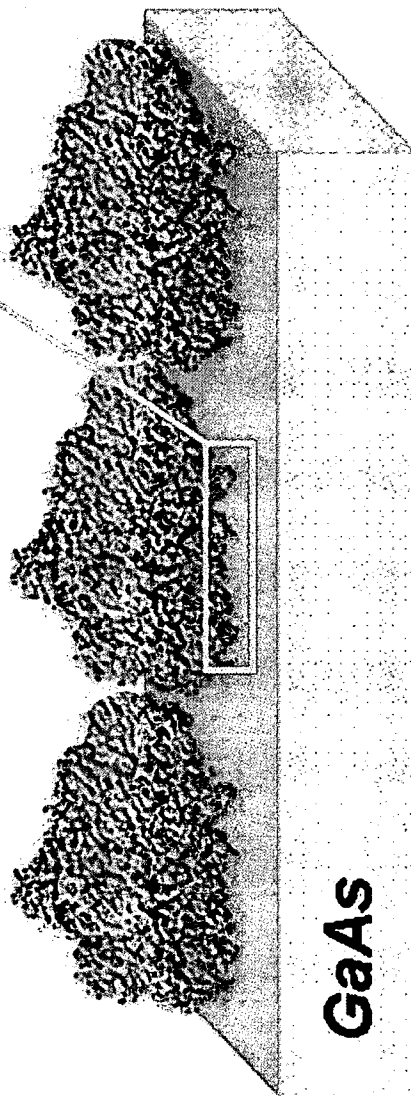


FIG. 1B

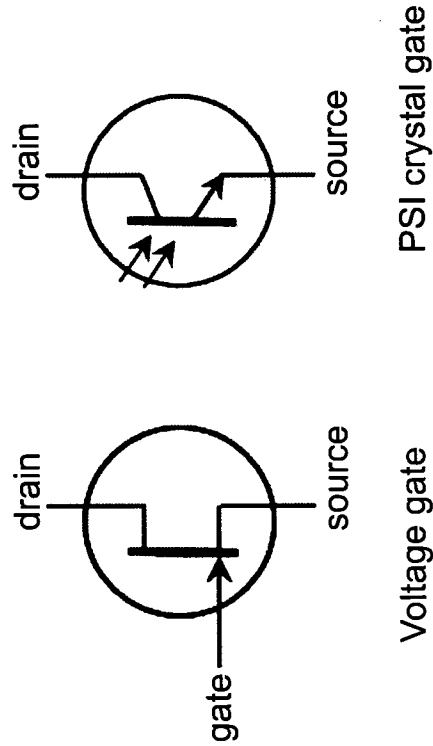
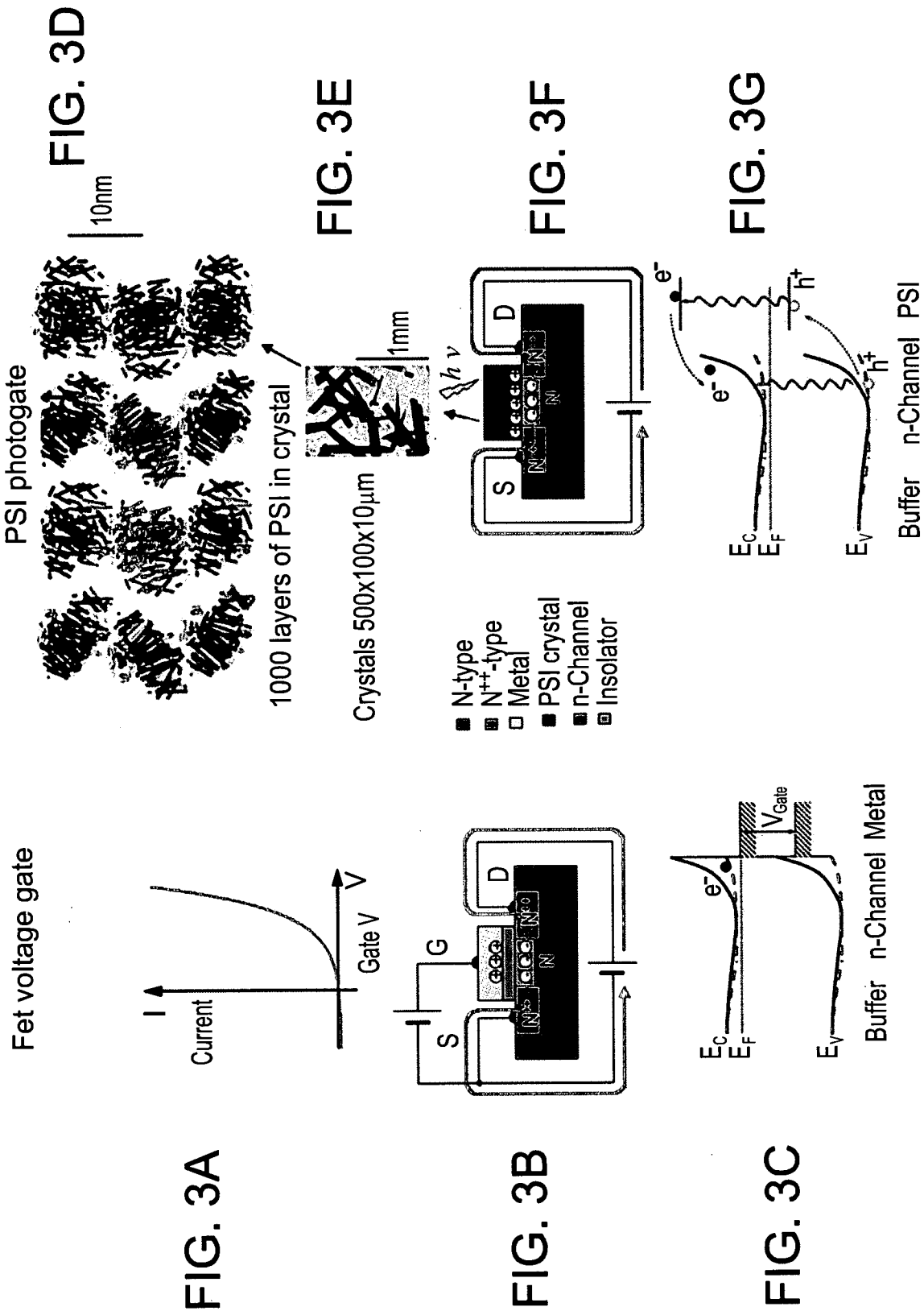


FIG. 2



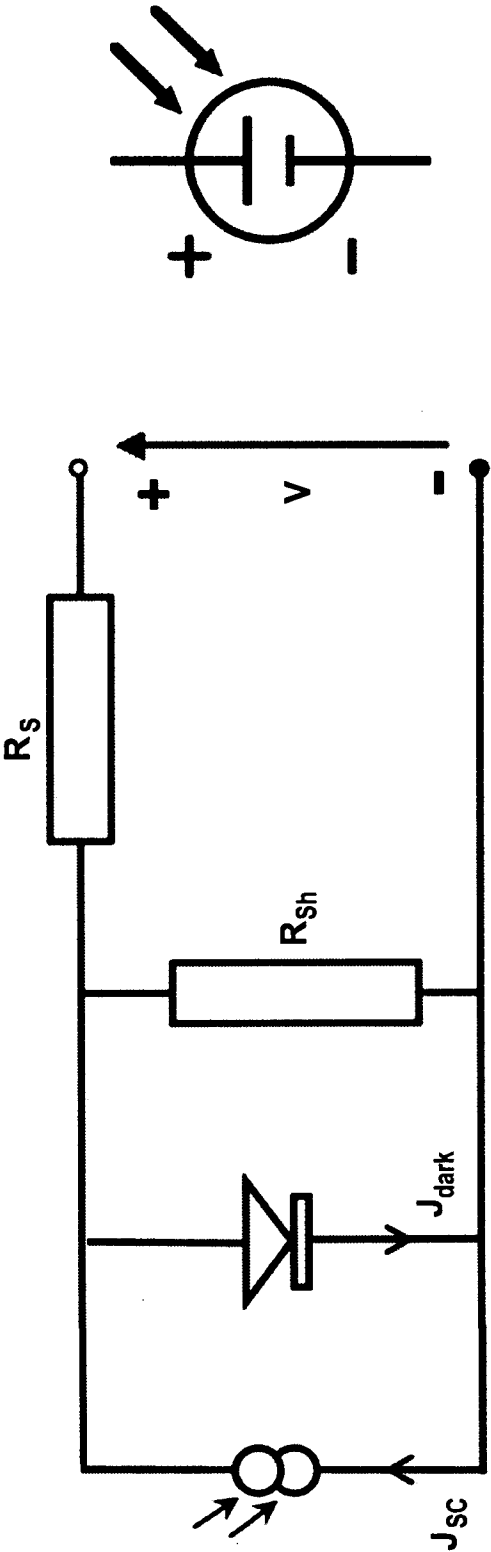


FIG. 4

5/13

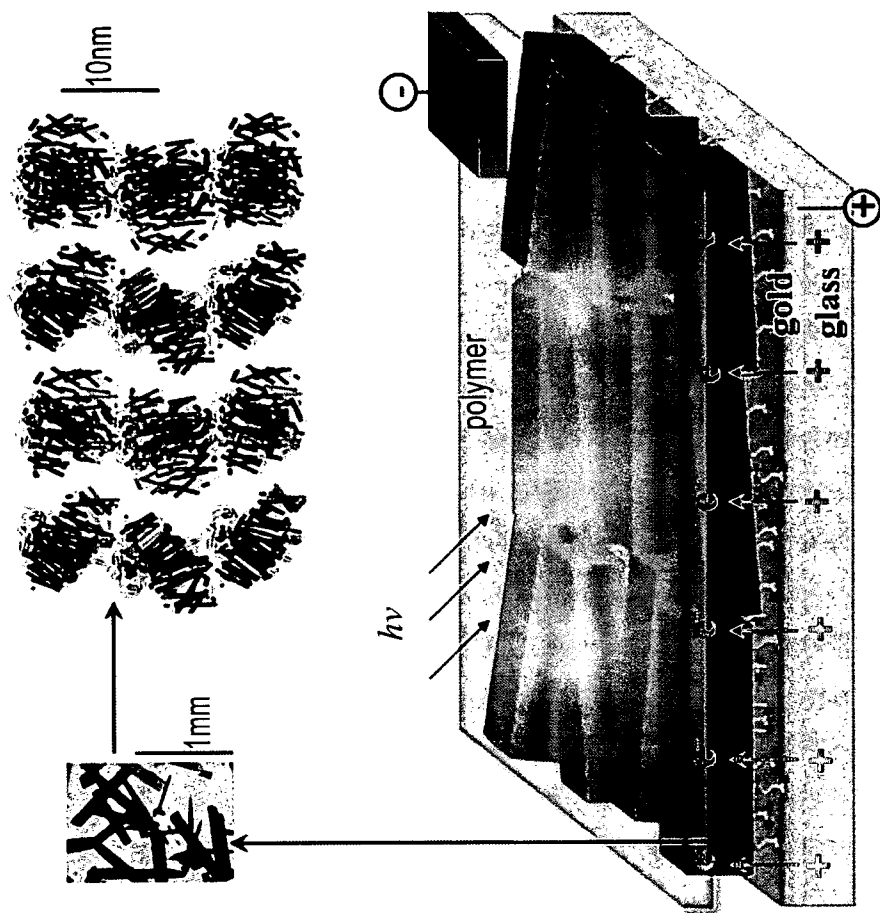


FIG. 5B

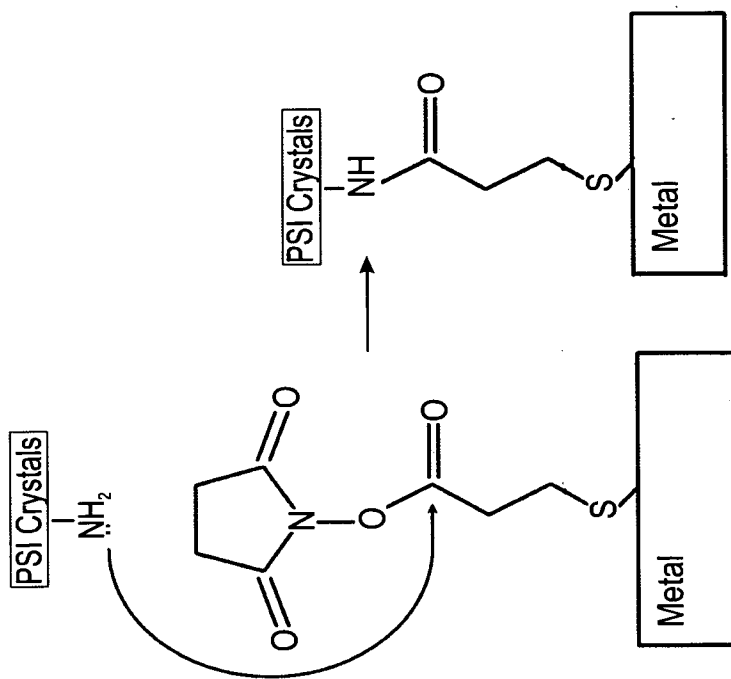


FIG. 5A

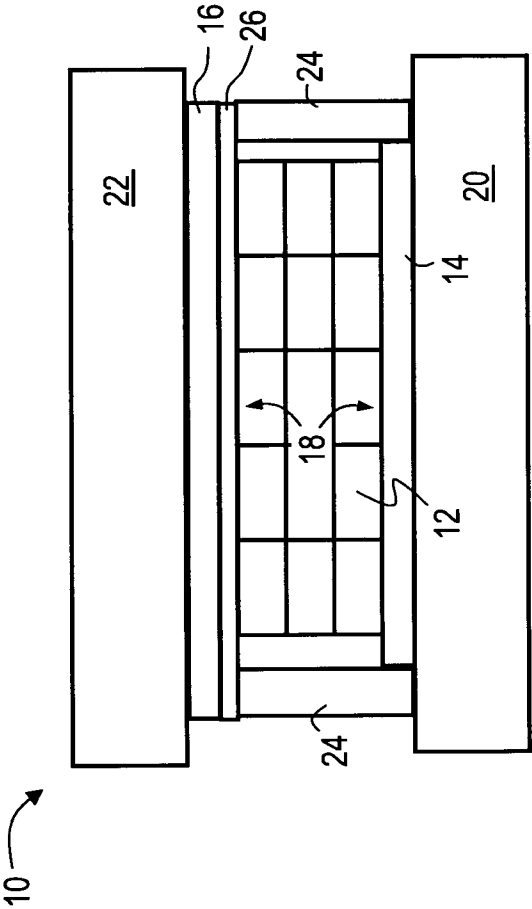


FIG. 6

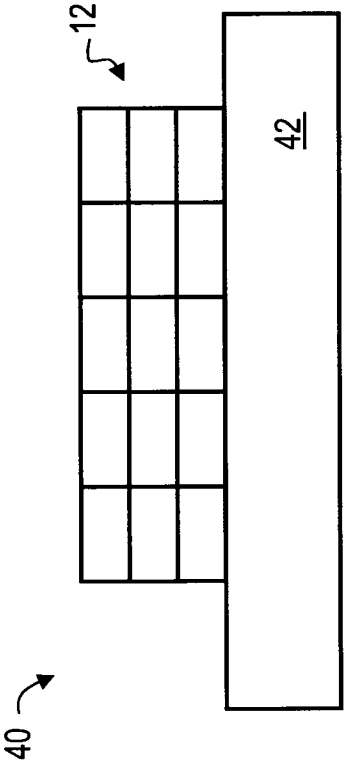


FIG. 7

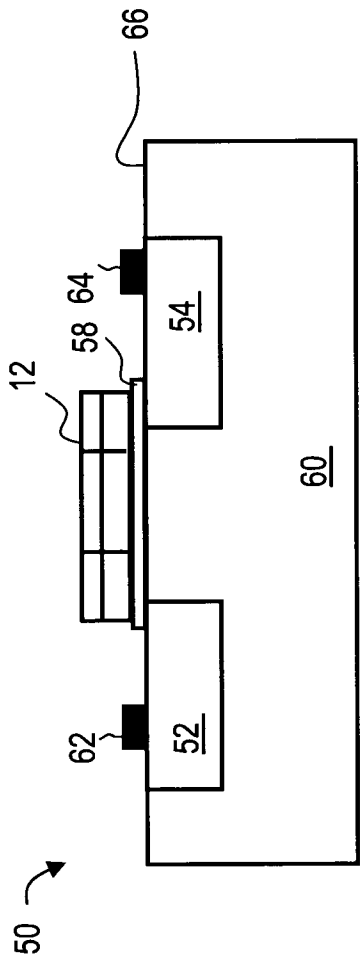


FIG. 8A

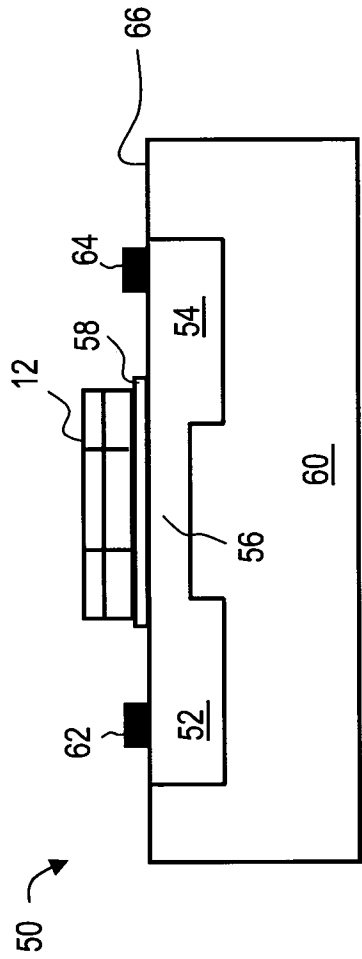


FIG. 8B

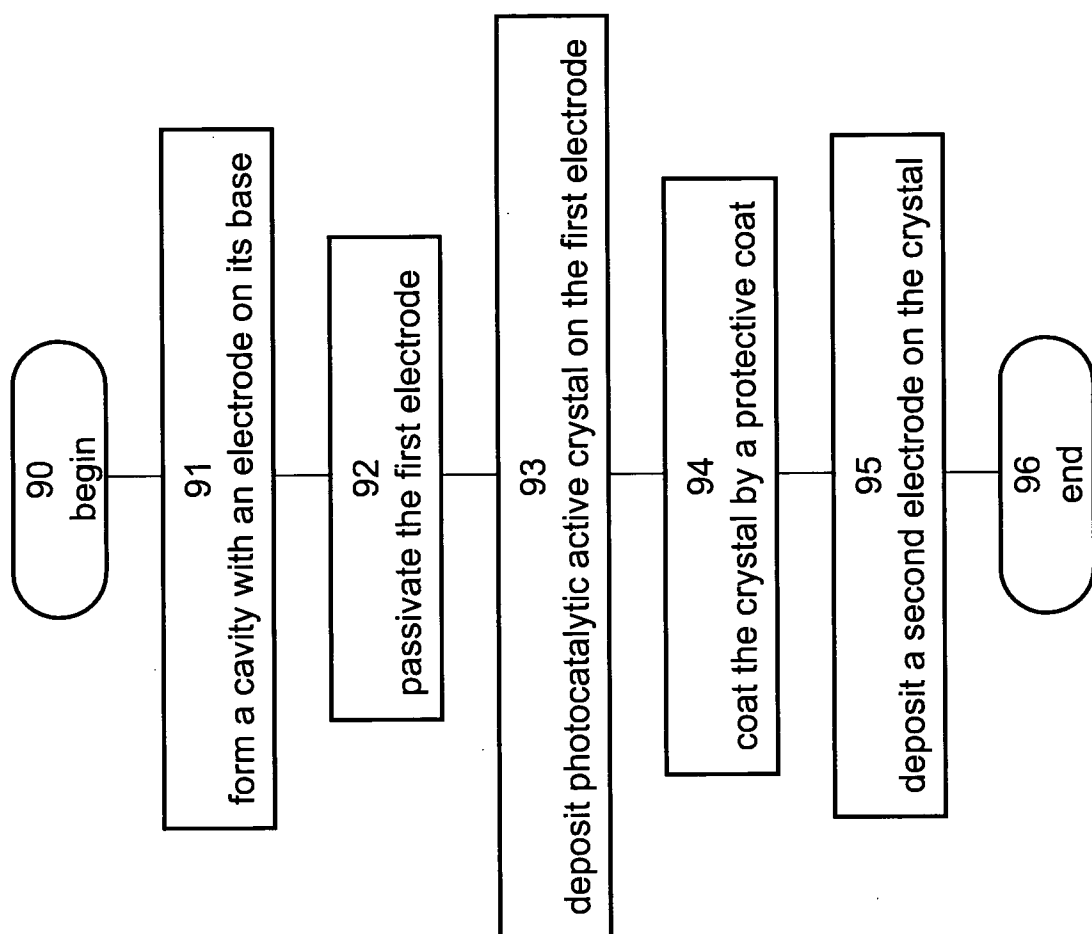


FIG. 9

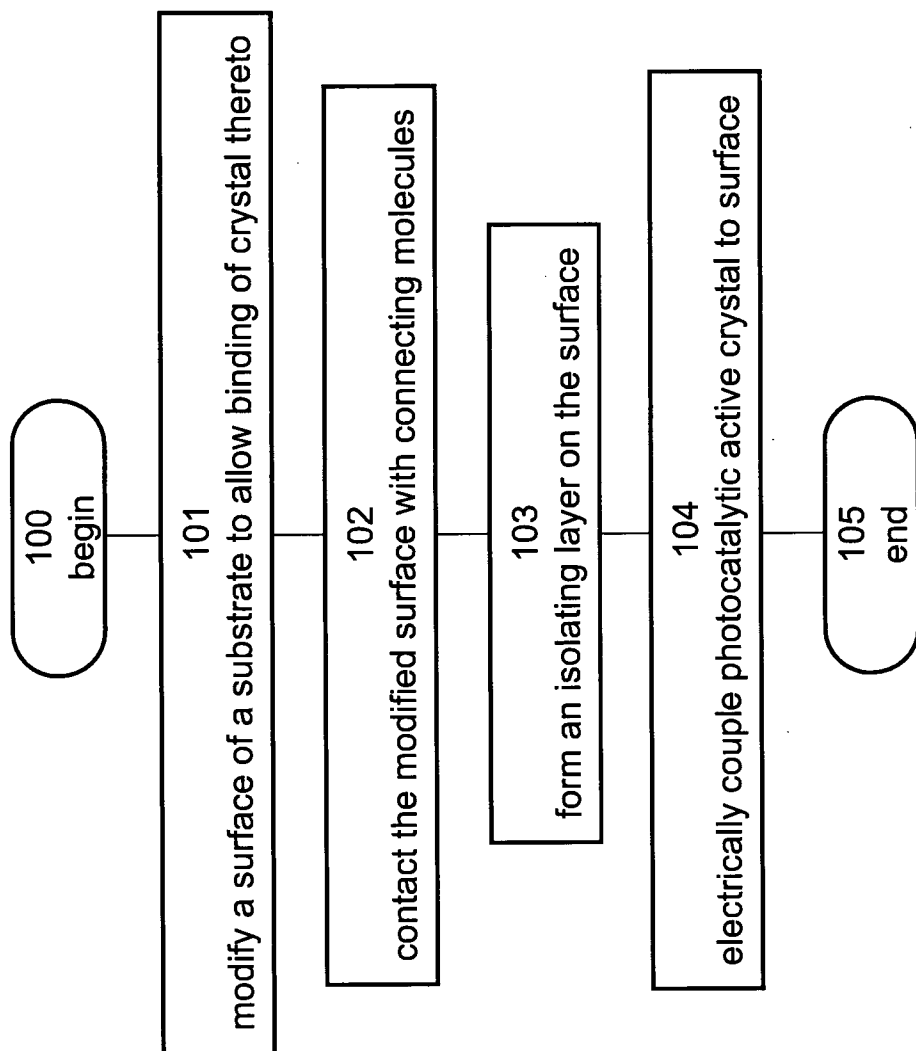


FIG. 10

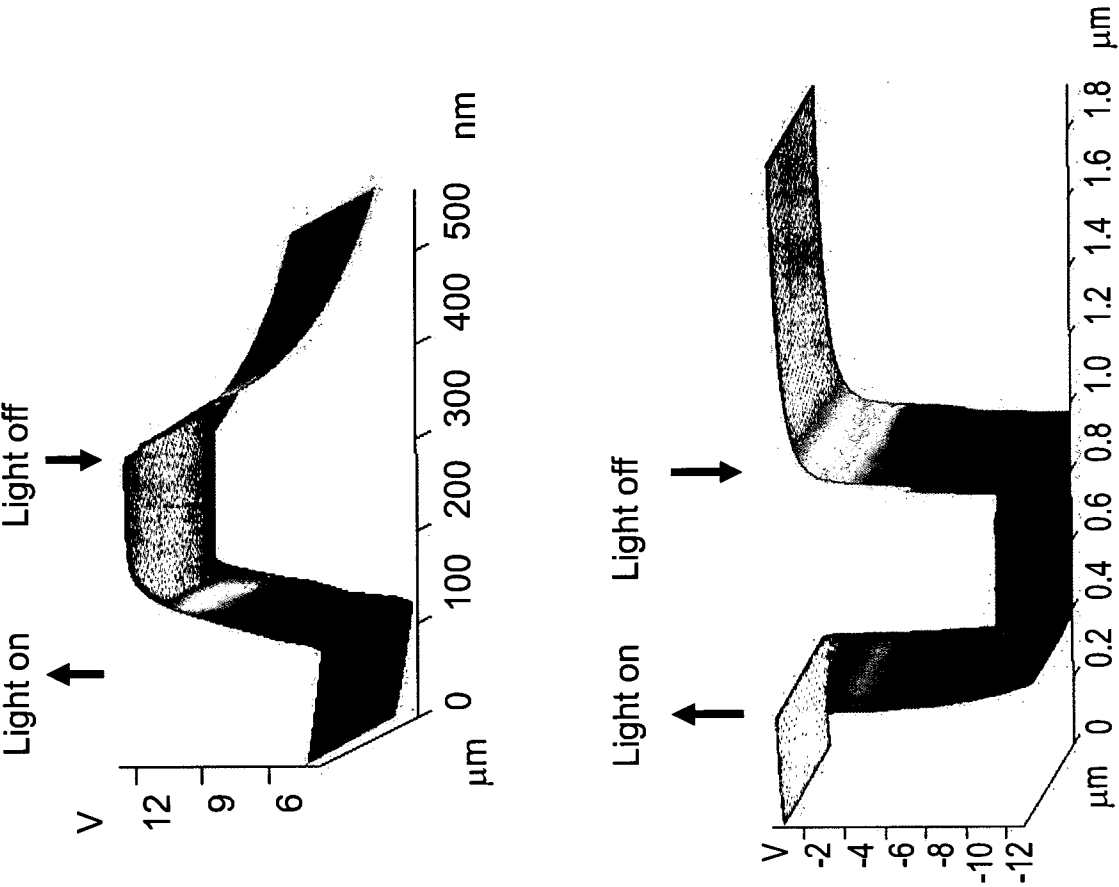


FIG. 11A

FIG. 11B

11/13

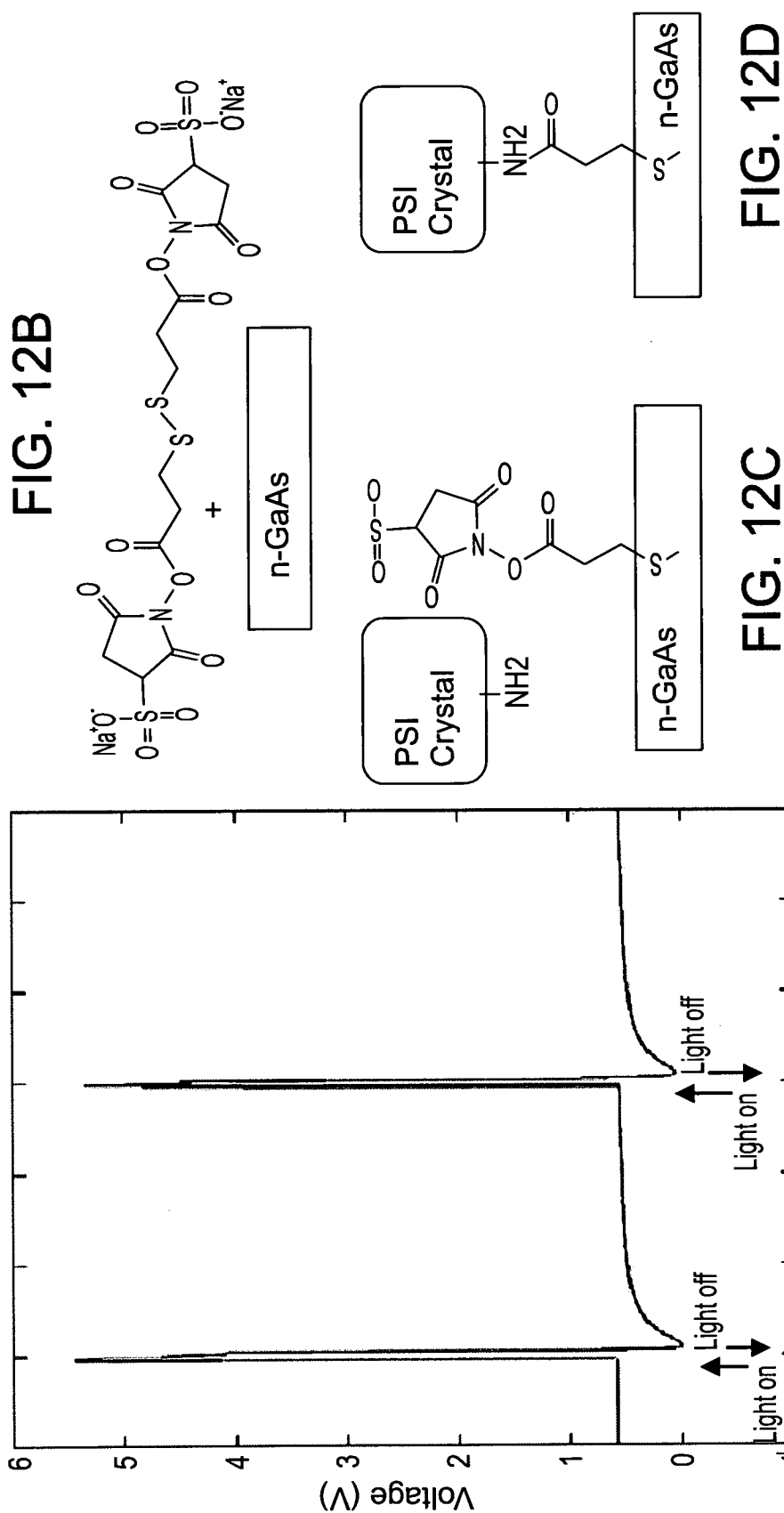


FIG. 12A

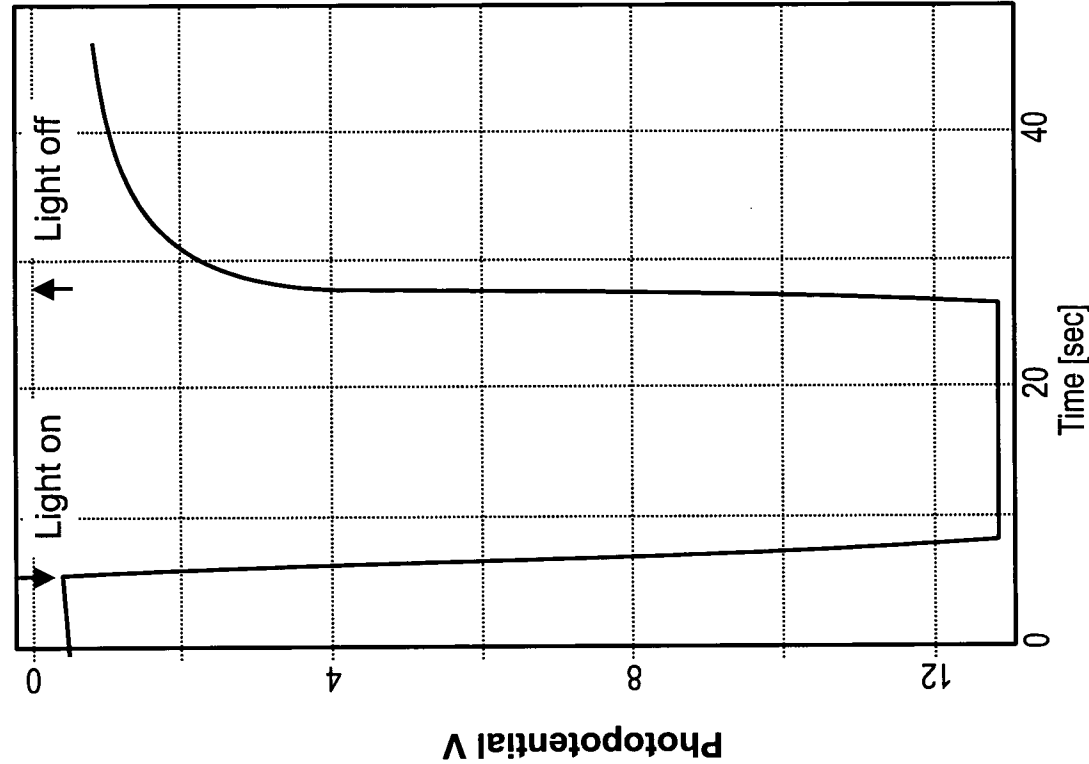


FIG. 13A

13/13

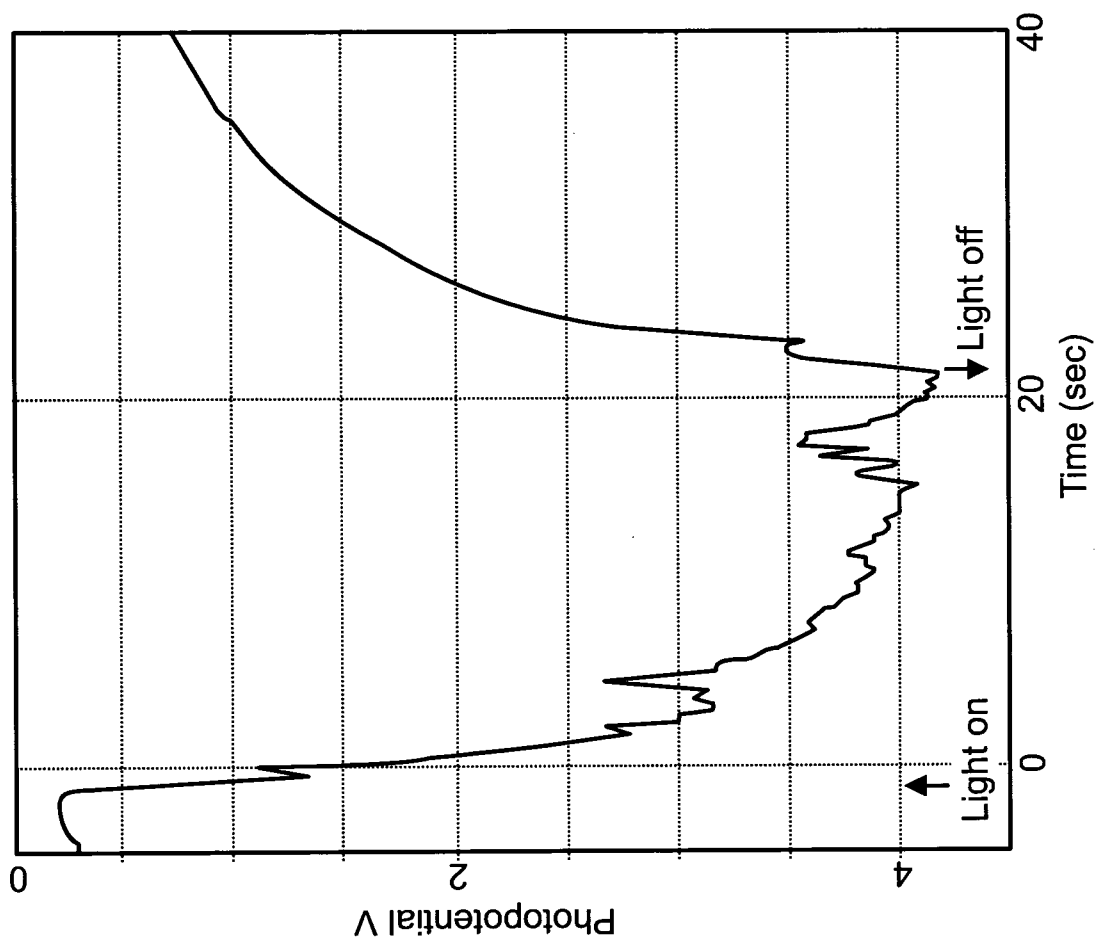


FIG. 13B

INTERNATIONAL SEARCH REPORT

International application No

PCT/IL2010/000412

A. CLASSIFICATION OF SUBJECT MATTER INV. H01L51/42 H01L51/05				
According to International Patent Classification (IPC) or to both national classification and IPC				
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) H01L				
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched				
Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, INSPEC, COMPENDEX				
C. DOCUMENTS CONSIDERED TO BE RELEVANT				
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
X	A. BEN-SHEM, F. FROLOW, N. NELSON: "Crystal Structure of Plant Photosystem I" NATURE, vol. 426, 11 December 2003 (2003-12-11), pages 630-635, XP002601469 the whole document	1,6-10, 16		
X	A. BEN-SHEM, N. NELSON, F. FROLOW: "Crystallization and initial X-ray diffraction studies of higher plant photosystem I" ACTA CRYSTALLOGRAPHICA SECTION D, vol. 59, no. 11, 2003, pages 1824-1827, XP002601470 the whole document	1,17-20, 23		
-/--				
☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.				
* Special categories of cited documents : <table border="0"> <tr> <td style="vertical-align: top;"> *A* document defining the general state of the art which is not considered to be of particular relevance *E* earlier document but published on or after the international filing date *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) *O* document referring to an oral disclosure, use, exhibition or other means *P* document published prior to the international filing date but later than the priority date claimed </td> <td style="vertical-align: top;"> *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. *&* document member of the same patent family </td> </tr> </table>			*A* document defining the general state of the art which is not considered to be of particular relevance *E* earlier document but published on or after the international filing date *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) *O* document referring to an oral disclosure, use, exhibition or other means *P* document published prior to the international filing date but later than the priority date claimed	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. *&* document member of the same patent family
A document defining the general state of the art which is not considered to be of particular relevance *E* earlier document but published on or after the international filing date *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) *O* document referring to an oral disclosure, use, exhibition or other means *P* document published prior to the international filing date but later than the priority date claimed	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. *&* document member of the same patent family			
Date of the actual completion of the international search 21 September 2010		Date of mailing of the international search report 26/10/2010		
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Königstein, C		

INTERNATIONAL SEARCH REPORT

International application No

PCT/IL2010/000412

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2008/023373 A2 (UNIV RAMOT [IL]; CARMELI CHANOCH [IL]; CARMELI ITAI [IL]; FROLOV LUDMI) 28 February 2008 (2008-02-28) page 6, line 17 - page 32, line 25; figures 1,10,11 -----	1,24-34, 40
X	WO 2006/060017 A1 (UNIV PRINCETON [US]; FORREST STEPHEN R [US]; PEUMANS PETER [US]) 8 June 2006 (2006-06-08) the whole document -----	27, 34
X	FROLOV L ET AL: "Fabrication of a photoelectronic device by direct chemical binding of the photosynthetic reaction center protein to metal surfaces" ADVANCED MATERIALS 20051017 WILEY-VCH VERLAG DE, vol. 17, no. 20, 17 October 2005 (2005-10-17), pages 2434-2437, XP002601471 DOI: DOI:10.1002/ADMA.200500295 the whole document -----	1,11-14
X	FROLOV L ET AL: "Fabrication of oriented multilayers of photosystem I proteins on solid surfaces by auto-metallization" ADVANCED MATERIALS 20080118 WILEY-VCH VERLAG DE, vol. 20, no. 2, 18 January 2008 (2008-01-18), pages 263-266, XP002601472 DOI: DOI:10.1002/ADMA.200701474 the whole document -----	1,4,5, 23,24, 27,28,34
X	FROLOV L ET AL: "Photoelectric junctions between GaAs and photosynthetic reaction center protein" JOURNAL OF PHYSICAL CHEMISTRY C 20080904 AMERICAN CHEMICAL SOCIETY US, vol. 112, no. 35, 4 September 2008 (2008-09-04), pages 13426-13430, XP002601473 DOI: DOI:10.1021/JP800586W the whole document -----	1,24-27

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/IL2010/000412

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 2008023373 A2	28-02-2008	WO 2008023372 A2	28-02-2008
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		JP 2008522428 T	26-06-2008