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fabricated today often have giant aspect ratios, readily exceeding the value of 1,000,000 – e.g. (Matovic & Jakšić, 2009). Such dimensions make them a hybrid between micro and nanosystems, even between macroscopic systems and nanosystems, since their lateral dimensions may be of the order of centimeters, while the thickness remains nanometric. Nowadays they are seen as a building block for various MEMS systems (Vendamme et al, 2006) , (Jiang et al, 2004a).

Since a biological complexity is sooner or later expected to be reached by micro and nanosensors and at the same time nanomembranes are the basic natural building block, it is only obvious to merge these two concepts into a single one.

In this chapter we show how such a simple fusion of two paradigms may result (and actually is already resulting) in a large multitude and variety of results. This is happening in spite of the fact that the field of nanomembrane-enabled sensors itself is extremely young, the first papers starting to appear several years ago (Jiang et al, 2004a). Here we consider only the application of synthetic/engineered nanomembranes and exclude biological structures.

After a concise overview of some of promising uses of nanomembranes in microsensors generally, we concentrate to a single sensor type, that of chemical, biochemical or biological (CBB) sensors utilizing the effects of adsorption/desorption and the surface plasmon resonance (SPR) effect. We consider the possibility to use nanomembranes as a platform for long range surface plasmons. The role of self-supported ultrathin structures in improving coupling between propagating modes and surface-bound plasmons is also analyzed, as well as their application in SPR sensor selectivity boost.

The ultimate in thickness of these building blocks is posed by the mechanical properties of the material itself, and the nanomembranes whose thickness can be of the order of several atomic or molecular monolayers certainly approach that limit.

Literature quotes the use of nanomembranes as the ultrathin freestanding structure to replace the conventional building blocks in deflection-based sensors (Jiang et al, 2004a). Among the obvious advantages of applying such a strategy are an increased sensitivity and a wider dynamic range. Various forms of micromachined freestanding ultrathin structures ensure much higher resonant frequencies than the conventional ones (extending into the GHz range).
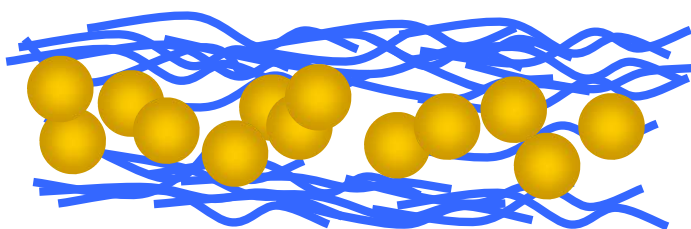
measurement of deflection in micromirrors, gyroscopes, etc. and structures in the thickness range 30 nm to 100 nm were fabricated in Si_3N_4 (Altena, 2006).

Another large field of application of freestanding nanomembranes are thermal sensors (Kruse & Skatrud, 1997). The need for large area thermal arrays of miniature detectors in infrared technology and remote sensing is large (Rogalski, 2003), (Dereniak & Boreman 1996). Various thermal detectors include bolometers (Richards, 1994), pneumatic detectors/Golay cells (Golay, 1947), (Chévrier et al, 1995), microcantilever-based devices (Datskos et al, 2004) to which bimaterial detectors belong (Djuric et al, 2007), etc.

Thermal detectors are typically based on a large and thin absorbing area which reacts to thermal changes due to its irradiation by electromagnetic radiation and is sensitive in the whole electromagnetic spectrum. Nanomembranes obviously offer smaller thermal inertia and thus promise faster operation and higher specific detectivities. The assessments of polymer nanomembranes with gold nanoparticle fillers in thermal detectors show sensitivities several orders of magnitude higher than those for silicon membranes with the same diameter. For instance, temperature sensitivities below 1 mK were calculated for 55 nm thick, 200 μm diameter nanomembranes (Jiang et al, 2004a). Nanomembranes freely suspended over microfabricated cavities dedicated to infrared thermal detectors were reported in (Jiang et al, 2006).

A large field of application of nanomembranes in (nano)photonics is their use in enhancement of the operation of semiconductor infrared detectors (Rogalski, 2003). These detectors are actually quantum devices whose operation is based on generation of charge carriers in semiconducting material upon illumination in a given wavelength range. Their sensitivity spectrum is much narrower than that of thermal detectors and its cutoff frequency is determined by the bandgap of the given semiconductor. One of the fields of the application of nanomembranes in such detectors is the fabrication of resonant cavity structures, which may be implemented as multilayer dielectric mirrors or one-dimensional photonic crystals (Jakšić & Djurić, 2004), (Djurić et al, 1999), (Djurić et al, 2001). In addition to their application as the building blocks for the resonator reflectors, such freestanding structures may be applied in devices with tunable resonant frequency, where electrostatic field is used to deflect the membrane and adjust position to furnish the desired resonant peak (Ünlü & Strite, 1995).

Another field of application are both tunable and fixed filters for photodetectors in various wavelength ranges obtained by lamination of planar structures (Jakšić et al, 2005), (Maksimović & Jakšić, 2006). There is also a possibility to modify and tune the emissivity and absorptance by the application of such multilayers (Maksimović & Jakšić, 2005), up to the point of creating thermal antennas for visible and infrared radiation (Maksimović et al, 2008).

Finally, a large field of application of nanomembranes is in chemical, biochemical and biological sensors based on plasmon resonance. The rest of this Chapter is dedicated to this important topic.

environment. The output is most often electrical or optical. The most important issues regarding a CBB sensor are its sensitivity and selectivity towards a given analyte.

A general CBB sensing system (Fig. 2) consists of three main blocks, (1) the unit for separation/filtering and possibly reaction enhancement, (2) the detection unit – the main part of the sensor where the signal is generated and (3) the processing unit where signals are conditioned and communicated further.
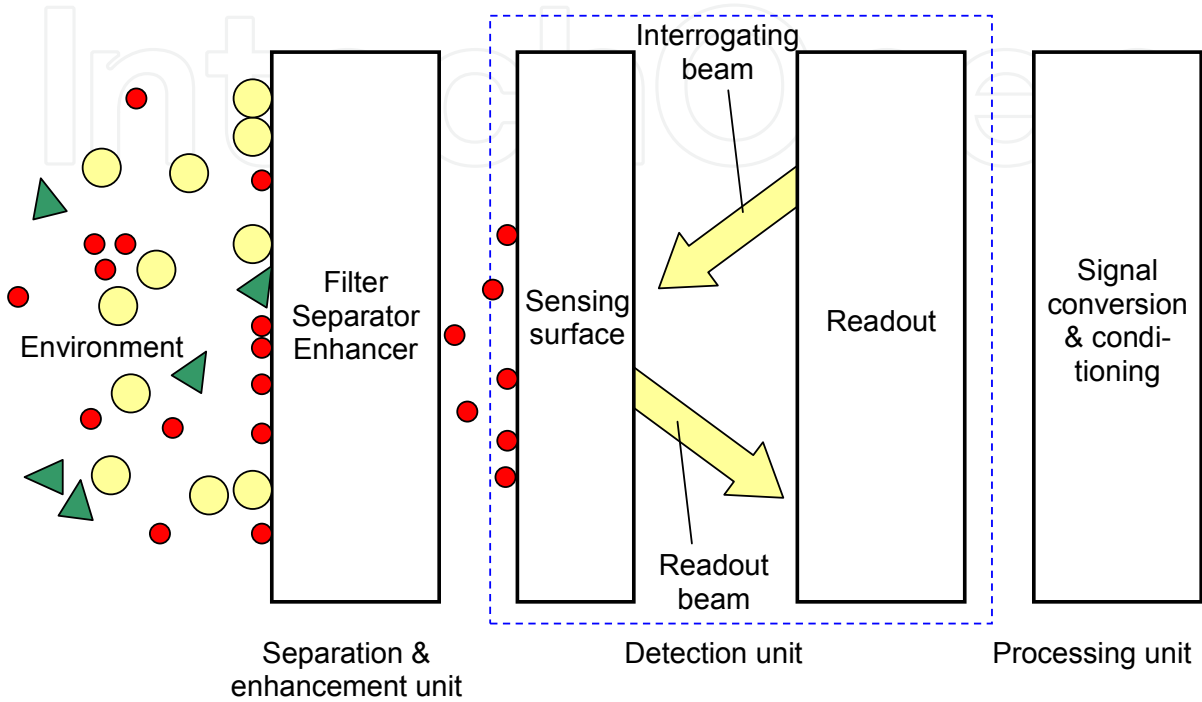

Fig. 2. Layout of a general CBB sensor consisting of (1) separation, filtering and enhancement unit; (2) detection unit and (3) signal conversion and conditioning unit.

We analyze the use of nanomembranes in the first two blocks. In the separation unit they are useful for filtering and generally molecular recognition if functionalized by nanopores, ion exchangers, absorbing fillers, etc., since their thickness enables a more accurate control of functionalization parameters than in larger structures. In the detection unit, especially of the kind used in nanoplasmonic devices, the nanomembranes are applicable as ultrathin, fully symmetric plasmon waveguides, strongly improving the device sensitivity.

analytes. Multichannel devices are readily implemented in such configurations. No moving parts are required and the fabrication technology is simple – the conventional SPP resonance-based sensor is a planar metal surface with the plasma frequency in the wavelength range of interest. Good metals are used to this purpose, typically gold or silver. Being fully optical, these sensors are resistant to external electromagnetic disturbances. Finally, plasmon sensors are very convenient for miniaturization and the fabrication of ultracompact sensor arrays.
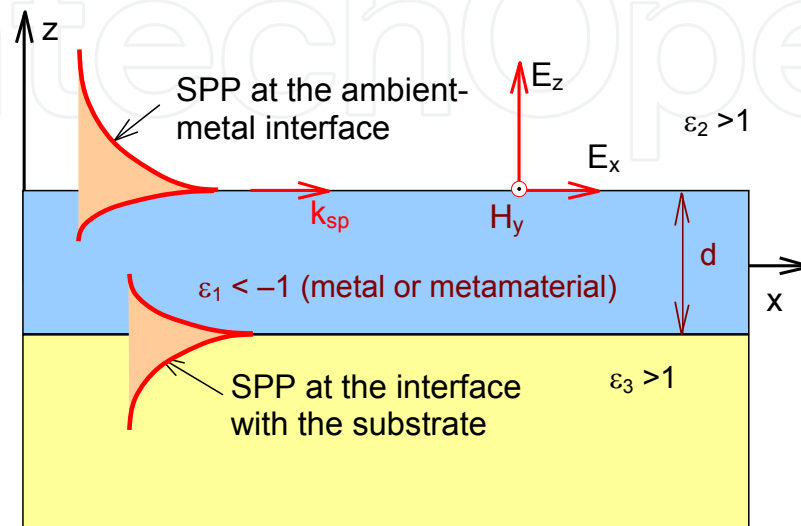

Fig. 3. Basic configuration of a guide for surface plasmon polariton propagation (metal-dielectric interface)

It is possible to use the same structure simultaneously for guiding SPP waves and for guiding the controlling electrical signals, since the active area is made of metal (Boltasseva et al, 2005). Also, the SPP components generally have high field localizations, thus promote the use of nonlinear photonic materials, ensuring the possibility for integration of active all-optical components (Zayats & Smolyaninov, 2003).

The operation of the SPP sensors is based on the modification of the propagation of surface plasmons polaritons at the sensor (metal)-environment (dielectric) interface. The analyte from the environment is bound either directly to the plasmonic surface, or (much more often) to a target-specific ligand layer. In both cases the surface refractive index is modified exactly in the position where the maximum of the SPP wave is located, since SPP waves are confined to the metal-dielectric interface and evanescent in perpendicular direction. In this way the maximum response is ensured. SPP resonance sensing is essentially thin film refractometry, where a change in the analyte concentration from c to $c + \Delta c$ causes a refractive index change at the metal-environment surface n to $n + \Delta n$ due to perturbed propagation conditions for the surface waves.

The obvious idea here is to use a nanomembrane as a waveguide for plasmons. Since a surface plasmon polariton is a quasi-planar electromagnetic wave decaying evanescently in both perpendicular directions, it is logical to utilize as a support for it a metal or metal-composite nanomembrane which is also quasi-planar.

Plasmons in nanomembranes with metal fillers were reported in (Jiang et al, 2004b), where gold nanoparticles were used in a polymer matrix and the packing density of the gold spheres varied from below 2% to about 25%. Experimental structures are typically light blue due to a plasmon resonance peak corresponding to the plasma frequency in visible.

Long-range surface plasmon sensors are especially convenient for biological sensors, since the confinement of the plasmon waves is smaller than in other SPP devices and thus the larger biological samples are more easily encompassed (Berini et al, 2008)

Probably the most important cause of the signal attenuation in LR SPP structures is its deviation from symmetry (Park & Song, 2006). Fig. 5 shows a calculated curve of attenuation for a metal nanomembrane immersed in dielectric.
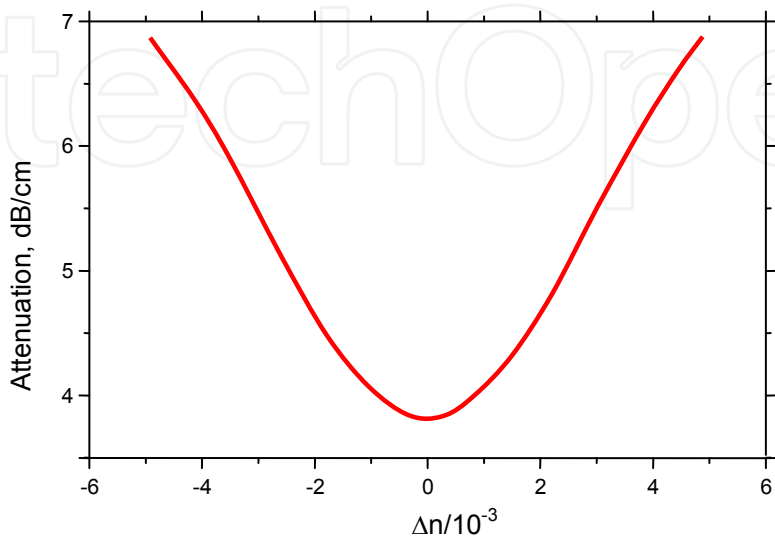

Fig. 5. Calculated LR-SPP propagation loss versus asymmetry of dielectric given as the refractive index difference. Membrane thickness 12.5 nm, material gold, refractive index of dielectric immersion 1.5, wavelength 1.55 μm.

It is visible that even very small deviations from symmetry introduce large losses into the waveguide.

The use of metal or metal-composite nanomembranes at the same time gives a platform for LR SPP and ensures its complete symmetry. Their thickness is typically from 4 nm up, thus very low losses are ensured. A layout of a nanomembrane-based LR SPP guide is shown in Fig. 6. The structure itself is extremely simple, being a freestanding planar nanomembrane sheet.
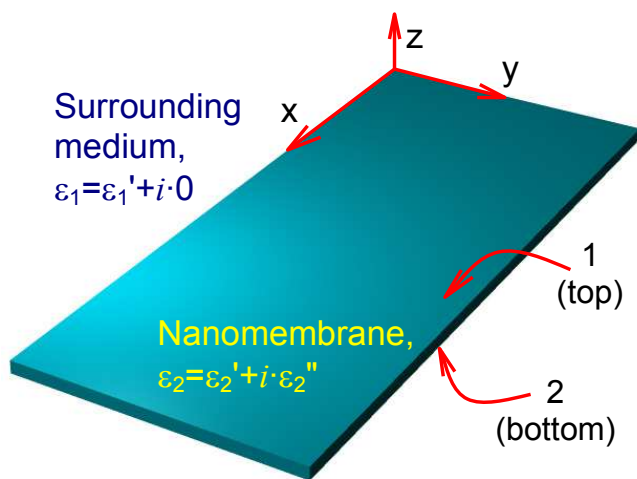

Fig. 6. Basic configuration of a freestanding nanomembrane guide for long-range surface plasmon polariton propagation (metal-dielectric interface)

The issue of coupling between the propagating modes and the plasmon waveguide is dealt with further in this text.

In coupling it is important to ensure that the maximum percentage of the incoming free-space mode is converted to SPP (and vice versa for the output). At the same time, it is important to ensure the smallest leakage and scattering losses. There are various schemes to ensure coupling between plasmonic devices and propagating modes. They may be roughly divided into four groups: prism couplers, endfire couplers, near-field probe couplers and those utilizing topological surface defects.

Historically the oldest methods are those utilizing prism couplers (Fig. 7). These include the Kretschmann configuration (Kretschmann & Raether, 1968) (Fig. 7a) which is still the prevailing readout method in plasmon sensors, as well as the Otto coupler (Otto, 1968) (Fig. 7b). Both of these methods utilize attenuated total reflection. Another method to excite the SPP is to use end-fire coupling, where the incident beam is in plane with the plasmonic surface (Fig. 7c) (Stegeman et al, 1983), (Berini et al, 2007).
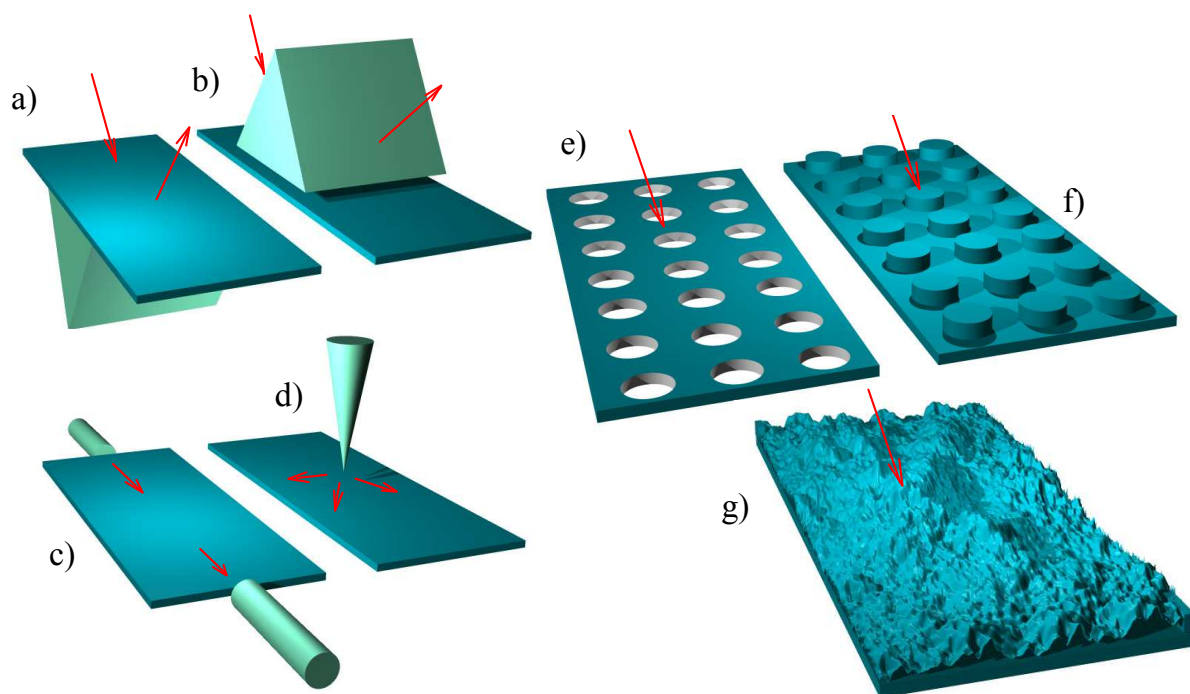

Fig. 7. Couplers plasmon-propagating a) Prism couplers in Kretschmann configuration; b) Prism couplers in Otto configuration; c) end-fire coupling; d) near-field probe excitation; Various methods of coupling through topological surface defects which may consist of e) gratings consisting of nanohole arrays, f) surface protrusions or may be g) disordered surface corrugations.

An important group of couplers utilize various near-field probes (the use of the "forbidden light' outside the light cone), (Fig. 7d) where local excitation in evanescent field is utilized and the beams tunnel from the impingement point to the metal-dielectric interface which supports SPP (Hecht et al, 1996), (Bouhelier & Novotny, 2007), (Maier et al, 2004).

Finally a large and very important group are couplers utilizing topological surface defects (Barnes et al, 2003), (Ritchie et al, 1968). These include grating couplers which may consist of periodic arrays of either subwavelength apertures (Fig. 7e) (Devaux et al, 2003) or surface protrusions (e.g. various pillars, bumps, etc.) (Worthing & Barnes, 2001), Fig. 7f. The arrays may be 2D like those shown in Fig. 7e, f) or 1D (grooves or stripes) and may have various shapes, e.g. rectangular, triangular, wavy, etc.

The couplers may be also disordered (this layout may be understood as a superposition of a large number of gratings with different periods) – Fig. 7g. (Ditlbacher et al, 2002)

In the case of freestanding nanomembranes and LR SPP sensors it is important to couple these structures with propagating modes with the least disturbance to the symmetry, thus preferably without a direct physical contact with the nanomembrane. One could use the shaping of a dielectric substrate (which, however, would perturb the electromagnetic symmetry of the structure), endfire coupling (which introduces alignment and coupling efficiency issues; it is known that the percentage of coupled light in this method is extremely low) or Otto prisms (bulky structure which makes the device significantly more complex).

We proposed an alternative approach which uses direct sculpting of the nanomembrane and is applicable without special alignment procedures (Jakšić et al, 2009). The idea of our approach is to incorporate the coupling structures into the freestanding nanomembrane itself, without any substrate to hold them. In this way the substrate and the superstrate remain fully index matched throughout the measurement. At the same time, the structure remains generally applicable, since the analyte does not have to be matched to the pre-fabricated device substrate.

The sculpted structures are small perturbations of the much larger nanomembrane, their dimensions being of the order of micrometers, while the membrane dimensions are measured in millimeters, even centimeters.

The surface is sculpted into a 2D array of protrusions (Fig. 8 a) which serve as a coupling diffractive grating (Kashyap, 1999). The basic approach to nanomembrane sculpting is illustrated in Fig. 8 b, c.
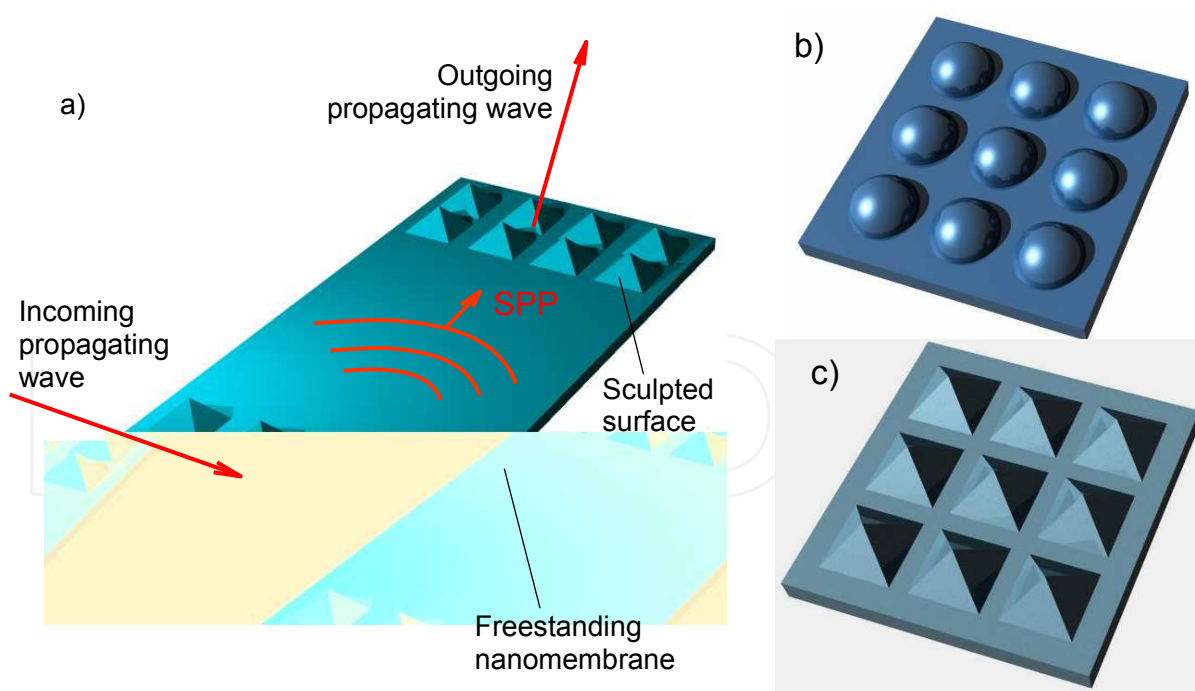

Fig. 8. a) Propagating wave to surface plasmon couplers using surface sculpting. b) Drawing of hemispherical surface relief for nanomembrane sculpting fabricated by isotropic etching through circular openings in photolithographic mask; c) Drawings of pyramidal surface relief for nanomembrane sculpting fabricated by anisotropic etching of silicon with (100) surface orientation through square windows aligned along [110].

The fabrication of surface-sculpted protrusion arrays is based on the deposition of a nanometric membrane precursor over a sacrificial layer (Mamedov et al, 2002), (Jiang et al, 2004b) and its subsequent release in etching solution, the same method used to produce metal-composite nanomembranes. The difference is that prior to depositing the membrane precursor, one first etches an array of micrometer pits in the sacrificial structure. The layout of the pits is determined by the applied photolithographic mask, thus defining the diffractive grating for the coupling. The shape of the pits themselves is defined by the chosen etching method. If isotropic etching is used, the pits are hemispherical or ellipsoidal. If anisotropic etching is utilized, one can produce for instance pyramids or truncated pyramids. The membrane precursor is subsequently deposited over the pits and upon release the fabricated nanomembrane retains the shape of the pits.

If the sacrificial layer pits are etched by isotropic etchant, the final protrusion are hemispherical, Fig. 8b. Variations to this generic form can be obtained by using different shapes of photolithographic masks (for instance, elliptical openings, but also various polygons) and by adjusting the etching duration to obtain either flatter or more voluminous structures.

The structure in Fig. 8c is obtained in an analogous manner, but using anisotropic etching of single crystalline silicon sacrificial layer with (100) surface orientation through square windows aligned along [110]. The variations to this generic form includes truncated pyramids and actually all standard forms obtainable by anisotropic etching.

Figure 9 shows the fabricated 15 μmx10 μm pyramid sculpted on the surface of a metal-composite nanomembrane with a thickness of 20 nm. It is known that nanomembranes become intrinsically stretched during their low-temperature annealing (Matovic & Jakšić, 2009) and thus the sculpted surface features retain their shape in spite of the minute thickness of their walls.
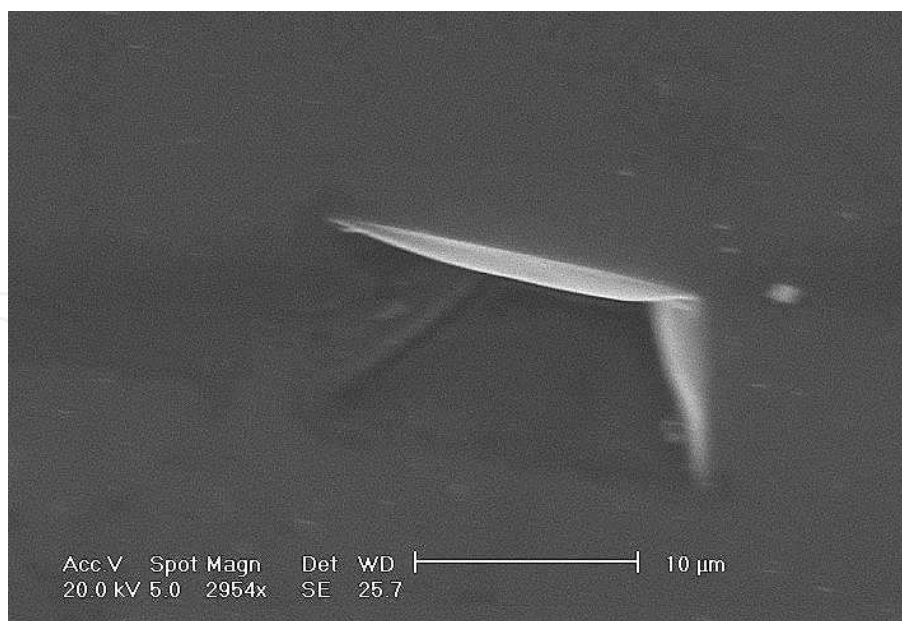

Fig. 9. Scanning electron microscope photo of a 15 μmx10 μm pyramid sculpted on the surface of a metal-composite nanomembrane, thickness 20 nm

We believe the tailorability makes the 3D surface-sculpted nanomembranes a valid alternative to other coupling methods for freestanding LR SPP sensor structures.
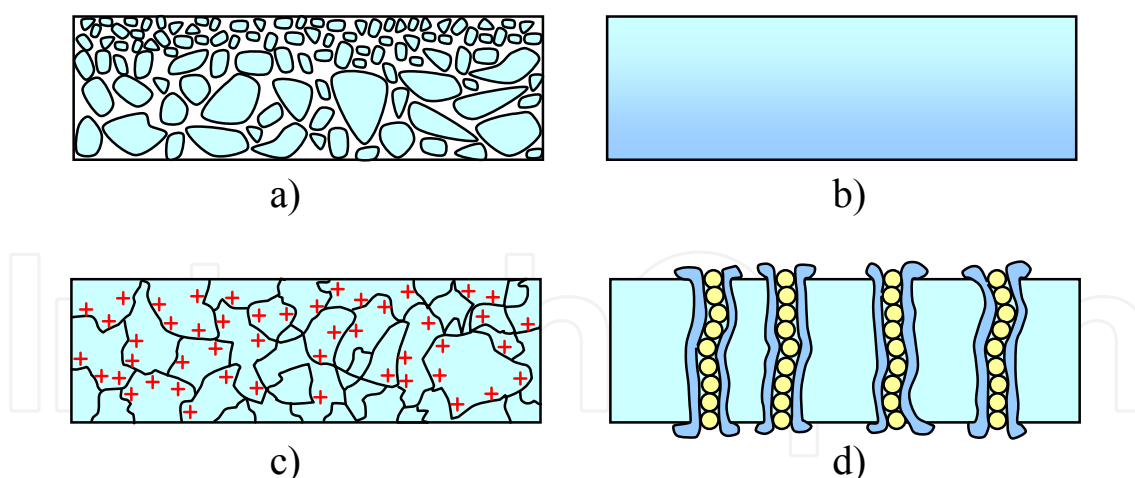
Fig. 10. Schematic presentation of selectivity enhancement through nanomembrane-enabled separation. a) Cross-section of nanomembrane with nanopores (Loeb-Sourirajan anisotropic structure); b) nonporous dense nanomembrane utilizing solution-diffusion mechanism; c) electrically charged membrane (ion exchanger); d) nanomembrane with gated ion channels.

2. Solution-diffusion through dense membranes where pores either do not exist or are at least smaller than the particle effective cross-section as defined by their thermal motion at a given temperature. The transport through such nanomembranes is a combination of particle solution and their diffusion across the structure, driven by concentration gradient, electrostatic field or pressure (Fig. 10b). These membranes can also be isotropic or anisotropic. They are used e.g. in pervaporation, reverse osmosis and often are regarded the method of choice in gas separation.

3. Ion exchange, where nanomembranes contain electrically charged particles (Fig. 10c). Most often such membranes are nanoporous, although they also may be dense. Positive or negative ions are fixed throughout the nanomembrane, usually at the pore walls. They bind the opposite ions and exclude the same charge. Structures with positive ions are denoted as anion exchangers, and those with negative ones are cation exchangers. This is the mechanism encountered in electrodialysis.

4. Gated ion channel flow, where the nanomembrane includes an ion-transmitting channel, typically consisting of proteins (Fermini & Priest, 2008) but which generally may be fabricated in various organic and inorganic materials. The channel is gated by external stimuli, typically by applied voltage or by ligands, which control the ion transport through the channel, Fig. 10d. This transport is highly selective. For instance, aquaporin proteins allow the flow of water molecules but are impermeable to protons, although these are much smaller than the water molecules. In other nanochannels transport of protons occurs, while the nanomembrane remains impermeable to other molecules (the Grotthuss mechanism, or proton hopping, where protons "hop" along a one-dimensional chain of water molecules – the "water wire") (Chung, 2007). Various ion channels exist for sodium, potassium ions, hydrogen, etc. This mechanism is fundamental for the ion transport through lipid bilayer nanomembranes in biological cells and represents the basis of the life on earth, while its biomimetic counterparts ensure another route to biosensor selectivity enhancement (Martin, 2007).

One can see that artificial pores are important for most of the described approaches. Nanopores are used to characterize DNA and RNA (Kasianowicz et al, 1996), up to the point of discriminating molecules differing by a single nucleotide (Vercoutere, 2003).

It is necessary to discern between different nomenclatures regarding the pore size, which may be the source of some confusion. According to the IUPAC, the term macropores is used to denote pores larger than 50 nm, mesopores have diameters between 2 nm and 50 nm and micropores are smaller than 2 nm (Rouquerol et al, 1993). In many literature sources all pores with diameters below 100 nm are termed nanopores (Aksimentiev et al, 2009).

There are several approaches to producing biomimetic pore complex capable of selectively transporting various biological analytes. Recently, an artificial scaffold for the nuclear pore complex-based gate was produced using natively disordered proteins termed FP nups. Such synthetic scaffold can be used as a generic platform for ultra-selective biomimetic nanopores (Jovanovic-Talisman et al, 2009). Another pathway is to avoid proteinaceous nanopores and to utilize other materials, including inorganic ones. Some materials used include silicon, silicon nitride, polyethylene terephthalate, silicon dioxide and many others (Aksimentiev et al, 2009). The design freedom offered by these approaches may lead to nanomembrane separators operating under less restrictive ranges of external parameters (temperature, electrolyte concentrations, pressures, pH values, bias, etc.). It is possible to tailor nanopores to practically any desired size, which means the freedom to optimize the pore geometry for various targeted analytes.

An important question is the method of combining the different parts of a CBB sensing device into a unified system. The unit for separation/enhancement may be integrated with the detection unit in various ways (Fig. 11). One may use two (or more) nanomembranes as separate elements so that the analyte-containing fluid is flowing sequentially through them, as shown in Fig. 11a. A modification of this approach is to utilize lamination of two (or more) separate nanomembranes into a single one, Fig. 11b. Finally, it is possible to aggregate detection and separation/enhancement functions into a single monolithic structure with one or more different active fillers which will perform affinity capture of the analyte and at the same time ensure tuning of the readout beam, Fig. 11c.
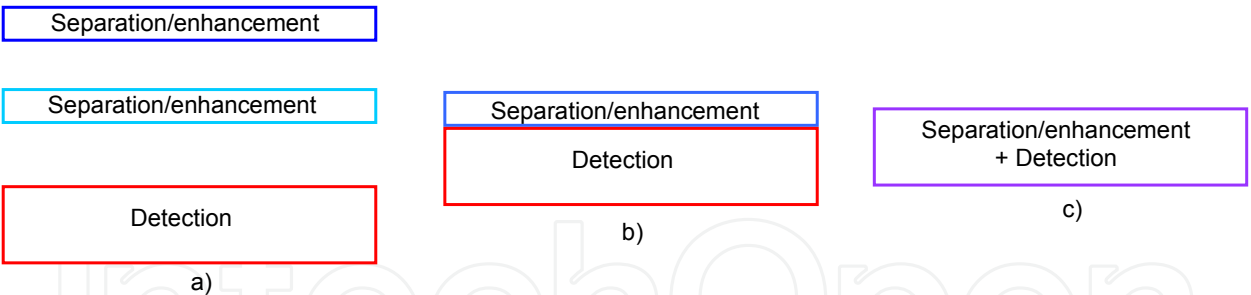

Fig. 11. Basic configurations of nanomembrane units for separation, enhancement and detection. a) The "separate separator" configuration where filtering structures are physically separated from the plasmonic waveguide; b) laminated structure, where separator/enhancer captures analyte; examples include e.g. solution/diffusion membranes, but also conventional ligand layers; c) aggregated/monolithic multipurpose unit.

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