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REVIEW ARTICLE

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Development of new classes of plasmon active nano-structures and their application in bio-sensing and energy guiding

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Metal nanostructures exhibit special optical resonance modes originating from the subwavelength confinement of conductive electrons in the material. These resonance modes represent a strong research focus due to their application potential in optics and sensing application. In this short review recent achievements of our group in this field are highlighted. A wet-chemistry approach synthesis of advanced metallic nanostructures will be introduced and their exact positioning and manipulation by electric field is shown. Next, the application of these nanostructures for a detection of small molecules will be described in several examples. Also, it will be shown that metal nanostructures can be used for sub-wavelength light focusing and for efficient energy coupling into polymer chains.

Keywords plasmonics, sensing, nanostructures fabrication, energy guiding

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Contents

1 Introduction

The specific optical properties of metals are determined by their loosely bound conductive electrons, which can be appointed as free electron gas in the material. Their large mobility and their plasma frequency lying at the ultraviolet range cause that visible light does not penetrate into the metal and is mostly reflected [1]. However, when the electrons are spatially confined into small volumes by creation of metallic nanostructures or interfaces then a coupled resonance state of light with collective oscillations of electrons, called surface plasmon polariton resonances (SPR), can be created at the surfaces of these metallic objects [2, 3]. The research field investigating these resonances is called Plasmonics. In general, plasmonics deals with generation, propagation and detection of these plasmon oscillations [4, 5].

These plasmon resonances were already observed in a large variety of metallic structures and new scientific articles describing other systems with specific properties are being constantly published. An overview diagram of the different geometries supporting SPR is given in Fig. 1. The shapes are ordered according to its complexity in form of a triangle, where the tips represent the basic SPR modes in zero, one and two dimension. A representative of zero dimension is a sub-wavelength metal nanoparticle. Such particle supports localized surface plasmon resonances [6], where the dipole mode is the most prominent. These resonances can be easily excited by direct illumination of the particles and their existence is confirmed by absorption or scattering measurement. In two dimensions a plane gold surface supports propagating surface plasmon modes [7]. These modes produce oscillating charges on the surface with evanescent light exponentially decaying from the surface. These modes cannot be excited by direct illumination, but excitation through a medium of increasing momentum of the illumination light is necessary. A plasmon on silver nanowires is a representative of one dimension and it can propagate along the wire [8]. These basic plasmon modes can be analytically expressed as solution of Maxwell equations [6, 7]. By deviating from the simple shapes or combining them, properties of plasmon modes will change and new plasmon modes can be created. These plasmon mixing and coupling is analog to an interaction of electron wave function of simple atomic and molecular orbitals [9] described by quantum mechanics. In cases, where the analytical solution is not available, various numerical methods such as Finite Element Method, Finite Difference Time domain, Method of Moments are used [10–13].

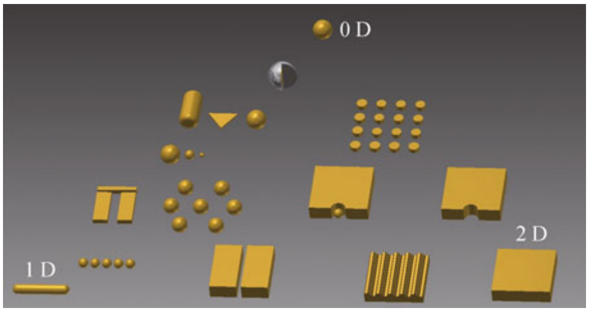

Fig. 1 A diagram of different metallic nanostructures supporting surface plasmon resonances. The geometries are distributed in form of a triangle, where the tips represent pure zero, one and two dimensional SPP modes and the geometries inside the triangle represents more complex modes or their combinations.

Metal nanoshells [14] are first example of extending simple spherical nanoparticles. The plasmon resonances of nanoshells can be nicely varied by the thickness of the metallic shell and can be tuned to the infrared light. Further, the nanoparticle can have non-spherical symmetry, such as rods [15], plates [16], triangles [17], etc. This enables the particle to have several different plasmon modes, where their spectral position depends strongly on the geometry. Another class is observed when the particles are close to each other so that the localized plasmon modes can interact with each other. This leads to a spectral shift of the single modes and strong localization of the modes between the particles [18, 19]. In this way a nanolense from spherical nanoparticles of different diameter are created [20]. Coupling of quadrupole plasmon resonance can be achieved by using a group of larger spherical nanoparticles [21]. This forms a strongly absorbing plasmon mode with a narrow bandwidth. A dolmen type structure [22] or radial symmetrically ordered nanoparticles around a central one [23, 24] can lead to the creation of Fano-type plasmon resonances, which is an interference phenomena of a narrow band plasmon mode spectrally overlapping with a broad band plasmon mode. These narrow modes are often dark field modes, which cannot be directly excited by light. Such interference can lead to an analog effect of electromagnetically induced transparency [25]. The plasmonic particles of different shapes can be ordered in non-periodical [26] or even densly packed periodical arrays [27].

The plasmon resonances in one dimension can be extended to propagating modes in metallic stripes [20], or grooves in metal layers [28]. The plasmon can also propagate in a chain of metallic nanoparticles [29]. In two dimensions a periodical pattern can be superimposed on a plain metallic layer. This creates a metallic grating [30, 31], in which the plasmon modes can be directly optically excited, because the extra momentum of the light is gained from the grating structure. Also a periodical nanoslits pattern in metallic layer can be produced [32]. This structure generates very sharp Fano type resonances in the transmission spectra. The compact metallic layer can be distorted by a sub-wavelength aperture [33] or concentric rings [34] leading to the enhanced transmission through the layer (extra ordinary transmission) caused by the generated plasmon of one surface and tunneling through the aperture to the other side. Finally, the nanoholes [35] or other perturbation [36] can be ordered in a periodic order to form structures exhibiting combination of propagating and localized surface plasmon modes.

There is a large variety of opportunities, where SPR can be implemented in various systems in order to improve them. The excitation of the SPR modes leads to a strong local electromagnetic field enhancement in the vicinity of the nanostructures and to a light absorption. This light focusing can be used to build antennas for visible light [37, 38] to effectively link propagating and spatially localized fields. Such spatially localized fields can be further guided on the nanostructures well beyond the diffraction limit of the light [39] enabling to scale down optical circuits. The light confinement is also widely examined for its possibility to increase a generally weak interaction of light with quantum systems such as molecules and quantum dots [40–42]. It can be either used to increase the light absorption by the molecules, application for example in solar cell technology [43], or to modify the fluorescence efficiency of the molecules [44]. This effect is used to increase efficiency of biosensors based on fluorescence [45–47]. An extremely strong enhancement of a signal is also observed in Raman spectroscopy (Surface Enhanced Raman Spectroscopy) for molecules close to metallic nanostructures [48, 49]. Further, non-linear optical effects can be supported and strongly enhanced in the metal nanostructures [50]. Recently, it was shown that special quantum properties of light such as entanglement and squeezing can be also detected in plasmons [51]. These experiments show a new direction in quantum plasmonics.

In the next section recent results in the field of plasmonic, which our group is working on, are presented in the following manner. In "Advanced synthesis of nanostructures" section, wet chemical-biological methods for synthesis of bi-metallic core-shell and "nano-flower" type nanoparticles are presented. It will be shown that the uniformity of the nanoparticle production can be significantly improved by using micro segmented flow synthesis. In the next section, methods for designed placement of nanostructures on substrates of such chemically prepared nanoparticles are explained. The following section shows how our group is exploiting these nanostructures for the detection of bio-molecules either optically or electrically. In the final part, the direct observation of light focusing into nanoscale volumes and coupling the light into energy guiding DNA strands is shown.

2 Results and discussion

2.1 Synthesis of nanostructures

The full potential of the plasmonics is limited by the ability to produce metallic nanostructures with sufficiently high precision and reproducibility. Comprehensive reviews on fabrication methods can be found in Refs. [52, 53]. In general, there are two technological routes, "top-down" and "bottom-up". In "top-down" physical methods are used. First, large continuous poly crystalline metallic layers are formed, which are locally removed by diverse methods such as photolithography, interference beam lithography, e-beam lithography or ion-beam etching. In "bottom-up" wet chemical synthesis are used. Metal atoms are aggregated in a controlled way to form nanostructures of desired shape. Our group is concentrated on "bottom-up" techniques.

2.1.1 Advanced core-shell nanoparticles

The preparation of metal nanoparticles using chemical synthesis is a well-established technique [54–56]. The material, material composition, shape and size of the particles can be adjusted by this step. These factors determine primary the plasmon resonances of the particles. The material composition can be realized in two defined ways. With two different plasmonic materials, like silver and gold, alloy nanoparticles and core-shell nanoparticles can be synthetized [57]. Especially, core-shell nanoparticles represent a fascinating field of plasmonics. In Ref. [58] we showed that the optical resonances of both metals can be finely adjusted by the chemically metal enhancement reaction [see Fig. 2 (a)]. The thickness of the shell compared to the dimension of the core determines the relation of both the silver and the gold-related plasmon resonances. For gold-cores with silver-shells the formation of hybrid resonances is also detectable.

Unfortunately, simple chemical synthesis does not allow a complex geometry for plasmonic constructions. Therefore, for the assembly of nanoparticles in defined arrangement molecular techniques based on biomolecular self-assembly can be used. These so called "molecular plasmonics" [60] show a large potential for future bio-sensor applications or as novel waveguiding elements. The principle of using molecules for nanoscale constructions are a long-standing idea [61, 62]. Particularly DNA conjugated metal nanoparticles and DNA conjugated surfaces can act as a building block for nanoscale constructions. The proposed nanoscale architecture can be realized by using highly specific biomolecule–biomolecule interaction, for example the hybridization of DNA strands. Nucleic acids are in nature in different conformations available, which can be exploited in nano constructions. Single stranded DNA act mostly as a "connector" between different components. Double stranded DNA can act as a 1D tube, for example as a conductive nanowire after a metal enhancement on the DNA. Also quadruplex DNA, the so called G-wires, are excellent building blocks for constructions [63]. These DNA molecules are more stable compared to double strand DNA. In case of DNA-based nanostructures, we can distinguish between pearl-chain-like 1D [64] and 2D constructions [65]. In the latter case, the comparison of different conjugation chemistry such as thiol- and amino-modification of the DNA molecules had a main effect on the stability and the building kinetics of the constructions. In the former case, specific arrangement between core-shell nanoparticles can be realized with the same method as constructions with pure metal nanoparticles, which we demonstrated in Ref. [59]. In Figs. 2(d) and (e) the scheme of the construction of nanoparticle pairs or flower-like particle constructions are shown and TEM images of the wanted NP complexes. These defined 2D-arrangements of plasmonic nanoparticles can be extended with novel DNA techniques for even more complex nanostructures, such as DNA-Origami [66].

2.1.2 Nanoparticle flow synthesis

There are many protocols for preparation of gold or

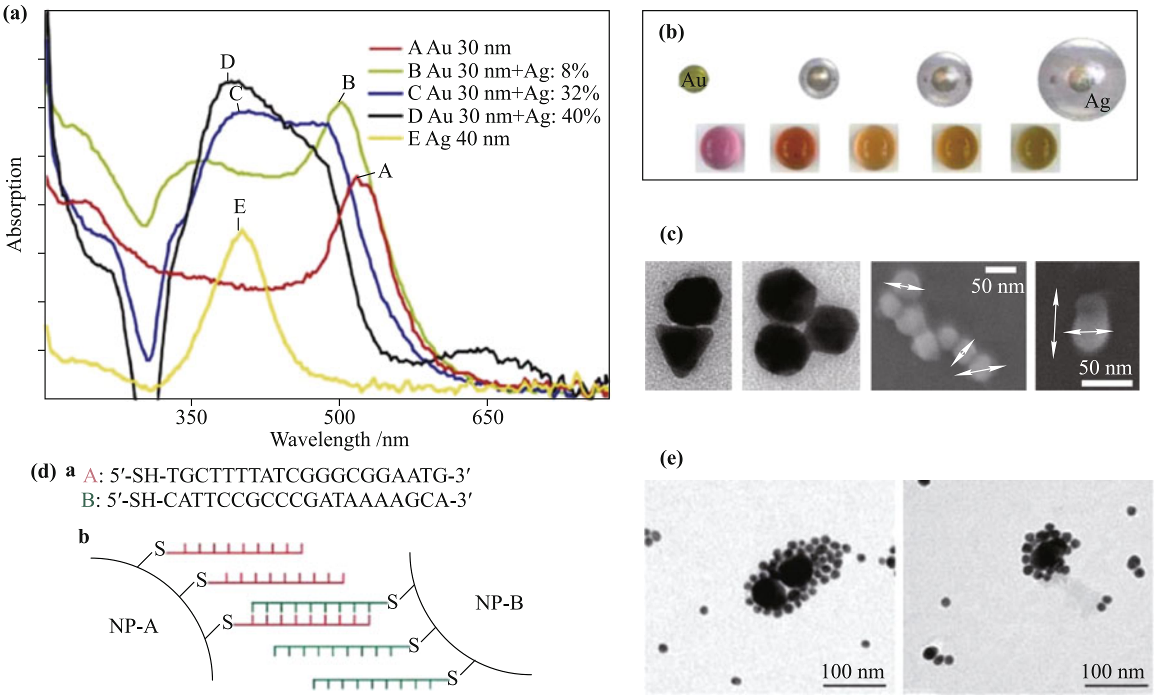

Fig. 2 **(a)** UV-vis absorption spectra of different types of gold core/silver shell nanoparticles. **(b)** Scheme of silver enhancement and photographic pictures of droplets containing gold core/silver shell particles. **(c)** TEM and SEM pictures of 30 nm gold nanoparticles enhanced with silver in solution. **(d)** Scheme of coupling of nanoparticles by DNA hybridization without the use of additional linker DNA. **(e)** TEM images of nanoparticles-DNA-nanoparticles constructs with different ratio of NP-A and NP-B. Reproduced from Refs. [58, 59] with kind permission from Springer Science+Business Media B.V.

silver nanoparticles by chemical reduction of metal ions in batch synthesis. The size and shape variation of the particles depends strongly on the local concentration of the reactants. Because in batch synthesis relatively large volumes are used, it is hard to achieve uniform concentration of the reactants in the whole volume. An elegant way of nanoparticle synthesis is applying microfluidics. It was shown in Refs. [67–69] that micro segmented flow synthesis can be successfully applied in production of various type of nanoparticle with narrow size particle distribution. The uniform distribution of reactants is achieved by mixing, which is extremely efficient in small volumes. A scheme of micro segmented flow synthesis is given in Fig. 3. The droplet formation is generated by continuous flow of two immiscible liquids: reactant solution and carrier medium. The nanoparticle synthesis is then initiated by injection of other reactants into the droplets. The NP synthesis in the droplets also drastically reduces the nucleation growth of the particles at the walls as in the case of batch synthesis, because the droplets are not in direct contact with the capillary.

It was demonstrated in Ref. [67] that core-shell and core-double shell nanoparticles with much better size distribution compared to standard batch synthesis can be achieved by this microfluidic approach. It was confirmed by comparing absorption spectra as well high resolution TEM images. In [68] this technique was extended to produce triangular shaped silver nanoparticles with tunable size and narrow distribution. The synthesis was carried out in two steps. In the first process, micro-segmented synthesis was used to produce spherical silver seed particles. In the second process, these seeds were mixed with a growth solution in droplets to create triangular shaped nanoparticles. The size of triangle is tuned by changing the number of seed particles in the droplets. The analysis of the particles confirmed higher quality of the nanoparticles prepared by micro-segmented flow than by a batch synthesis. The microfluidic methods also reduce the risk of aggregation of the particles.

A significant advantage of segmented flow synthesis is immediate characterization of the particles during the synthesis and flexibility in the adjustment of the conditions of the synthesis. This allows automatic computer controlled screening through the large space of parameters such as concentration of reactants, time of reactions and flow rates in order to obtain ideal conditions for nanoparticles synthesis, which was demonstrated in [67].

2.2 Nanostructures manipulation

The chemical synthesis of nanostructures has the

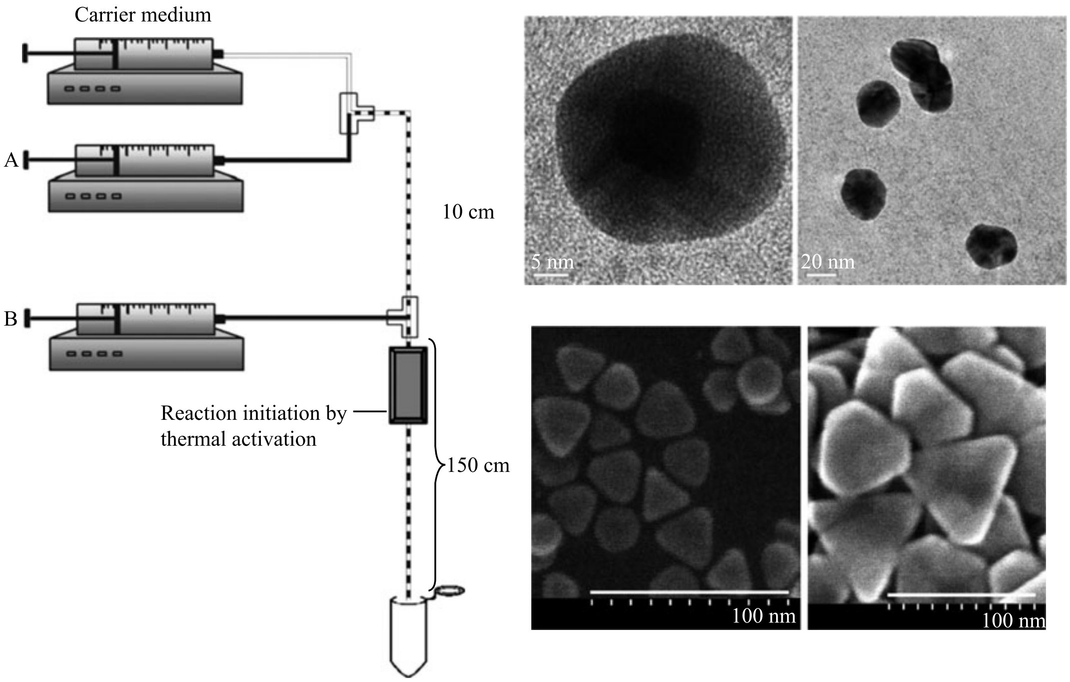

Fig. 3 *Left*: General set-up for micro segmented flow synthesis of bimetallic multishell nanoparticles. *Right*: HRTEM pictures of Au/Ag core/shell. *Right bottom*: SEM images of triangular shaped nanoparticles with different sizes. All these NPs were prepared by micro-segment flow synthesis. Reproduced from Ref. [67] with permission from Elsevier and reprinted from Ref. [68] with permission from American Chemical Society.

advantage of creating large amounts of the nanostructures in parallel dispersed in a solution phase. There are many applications, where the solution phase of nanoparticles is desired. In this case, the nanoparticles are mostly used as labels [70] or for a detection based on aggregation [71, 72]. However, in many cases it is desirable to immobilize the objects on specific places on a surface, in order to interrogate (optically, electrically, thermally, etc.) them.

Our group has long experience with controlled assembly of nanostructures and molecular chains on complex substrates. In our recent publications [73, 77–79] it was demonstrated, how metallic nanoparticles can be immobilized inside micro-structured optical fibers (MOF). MOF consists of various arrangements of micro-scale air holes in solid glass cores, and by engineering of such arrangement desired spectral and spatial characteristics of the guided light can be achieved. A SEM image of such fiber profile shows Fig. 4(a). The light is concentrated around the central part of the inner side wall of the capillaries. Therefore, the nanoparticles had to be attached inside the MOF. A dynamic low pressure deposition of metal NPs, the so-called nanoparticle layer deposition (NLD) method, in which NPs are chemically attached in a self-assembled monolayer, was used. First, a silane solution (APTES) was pumped through the MOF, which binds on the inner glass surface and acts as a binding site for the nanoparticles. In the second step, nanoparticles are pumped through the MOF. The density of immobilized NPs is in saturation coverage. This method allows extreme homogenous nanoparticle coverage along some meter fiber pieces. A major challenge was to reproducibly flow the solutions through very long but narrow capillaries in MOF and it was achieved by utilization of specially designed microfluidic chips for fiber-fluidic coupling. In this way, it was possible to uniformly cover up to 6 m long fiber with holes of 30 micrometer diameter.

It is also possible to specifically place small objects into micrometer size gaps between two electrodes [see Figs. 4(d), (e)]. The principle is based on Dielectrophoresis (DEP) [80], in which a force acts on a polarizable objects in non-uniform electric field. In practice an alternative voltage is applied on electrodes, which creates the non-uniform electrical field between the electrodes. Then objects are either pulled towards or away from the biggest gradient of the electric field depending on the polarisability of the solution and the objects. In this manner we successfully stretched DNA molecules [81] or silicon nanowires [75] between the tip of electrodes. The electrodes for catching the silicon nanowires had specially designed tapered shapes, which increased efficiency of the trapping. At the initial phase the nanowires were pulled to the electrodes and due to large size, this process was quite efficient. In the second phase, the nanowires were moved towards the end of the electrodes because of the

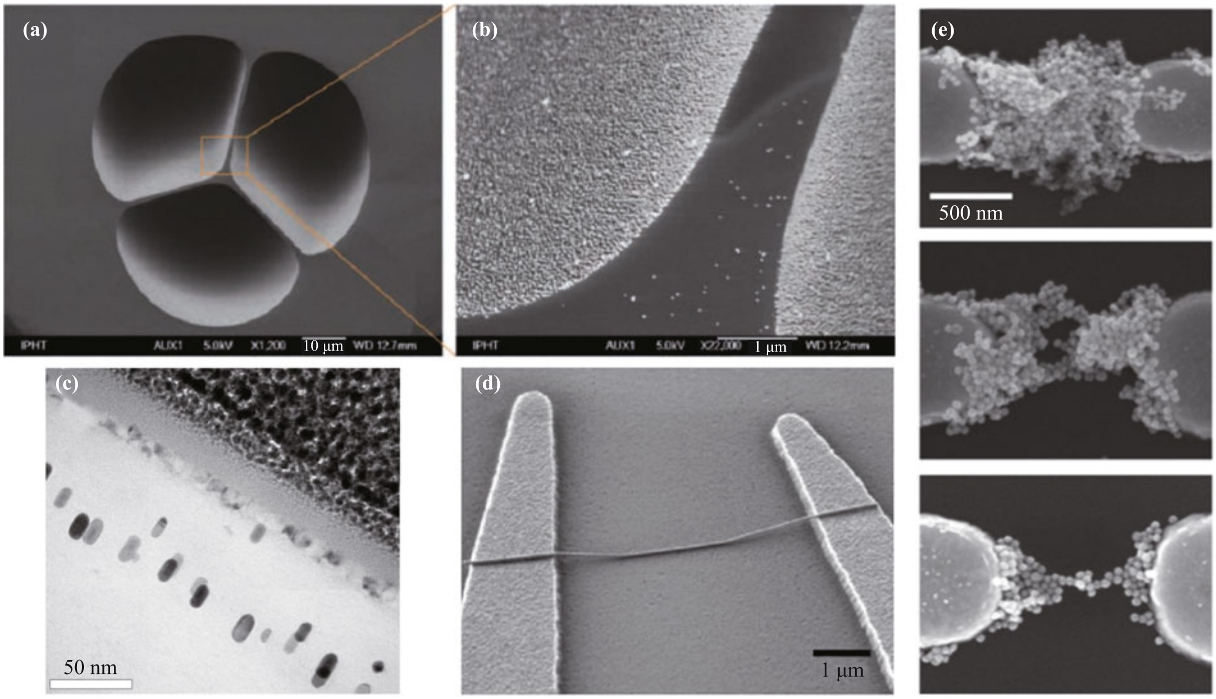

Fig. 4 **(a, b)** SEM images of the inner walls of microstructured glass fibers (MOFs) internally coated with gold particles (30-nm-diameter spheres). **(c)** TEM micrographs of characteristic alignment of gold nanoparticles within a strontium titanate (STO) matrix, here with a film thickness of 140 nm. **(d)** SEM image of dielectrophoretically (DEP)-trapped SiNW. **(e)** AuNP-chains with particle diameter 60 nm have been assembled in electrode gap under different conditions (1 MHz, 5–40 min, 1–2.5V). The voltage increases and the time decreases from top to bottom row. Reprinted from Refs. [73, 74] with kind permission from The Optical Society, from Ref. [75] with kind permission from Springer Science + Business Media B.V., from Ref. [76] with kind permission from Elsevier.

electric field gradient along the electrodes. Finally, nanowires were fixed by applying a short pulse of higher voltage, which improved electrical contact between nanowires. This welding step was an important improvement, because then it was possible to measure electrical properties of a single nanowire. Since such nanowires have also interesting optical properties [82, 83], the opto-electrical properties of individual nanowires could be measured in this set-up as well.

We also used this dielectrophoretic principle to immobilize metal nanoparticles between the electrodes and to create chains of nanoparticles [76]. It was shown that the morphology of the nanoparticle bridges can be varied by changing the nanoparticle size, applied voltage and the time. Example of metal NP bridges are seen in Figure 4 e. This assembly process requires only low cost equipment and can be done on many electrodes simultaneously.

In Ref. [74] an alternative route of production oriented anisotropic nanoparticles was published. This method relays on self-organization of metal nanoparticles from metal layer by elevated temperature, which is embedded in a $SrTiO_3$ matrix. The matrix induces the formation of gold ellipsoidal particles and they are aligned perpendicular to the substrate with its longer axis. The TEM image of such nanoparticles is presented in Fig. 4.

2.3 Application of nanostructures in bio-sensing

2.3.1 Sensing with Localized Surface Plasmon Resonance

The most exploited feature of SPR for use in molecule sensing is the SPR spectral sensitivity on a change of polarisability (refractive index) in the vicinity of the plasmonic structures [6, 7]. This principle is utilized in a construction of label free detectors for bio-molecules [84–93]. An adsorption of molecules on the plasmonic structures changes the local polarisability, and therefore, the frequency (or equivalently spectral position) of the SPR is moved. The shift of SPR is monitored by optical spectroscopy. The specificity of the sensors is based on immobilization of bio-recognition element (proteins, peptides, antibodies, complimentary DNAs, aptamers, polymer matrixes, etc.) on the structure, to which a molecule of interest (analyte) selectively binds. The major advantages of this technique is the possibility of real time measurements, the high degree of parallelization, enhanced sensitivity due to the strong confined field and system integration capabilities.

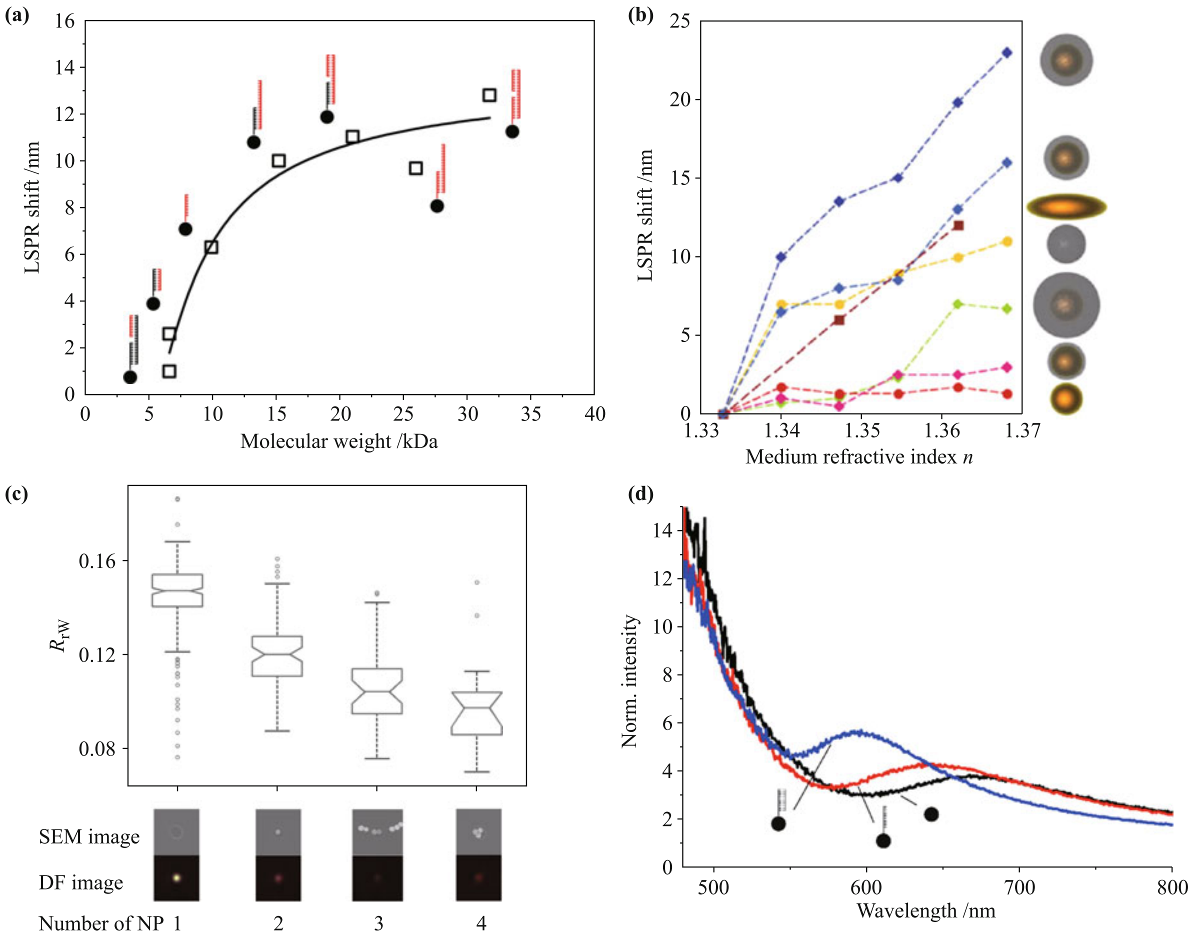

Fig. 5 **(a)** Non-linear dependence of a change in molecular weight by hybridization of DNA strands (red) to the resulting shift of the plasmon resonance peak. It shows the localized spatial resolution of LSPR sensing. **(b)** Experimental comparison of the sensitivity of different types of nanoparticles against medium refractive index changes and confirming the high sensitivity of core-shell nanoparticles. **(c)** Box plot of R_{rW} parameter for various particle populations. R_{rW} parameter is obtained from camera images of the holes only. *Insets*: Microscopic images (SEM, optical transmission) of a hole without and with particles inside. **(d)** Demonstration of specific DNA detection by a nanoparticle-nanohole arrangement: The spectra show that binding of capture DNA on the particle leads to a certain blue shift (more than 20 nm) of the dip at about 600 nm wavelength. Incubation with analyte DNA results in binding of the analyte to the capture DNA (hybridization) leading to a further blue shift of about 25 nm. Reproduced from Refs. [95, 94] with kind permission from Springer Science + Business Media B.V., from Ref. [97] with permission from American Chemical Society.

Our group's particular interest is in DNA detection on a single particle level. In a general experimental arrangement, the nanoparticles are immobilised on glass substrates, observed through an optical microscope in a dark field setup and individually spectrally measured by a microspectrometer. The ability of short DNA molecules to form a self-assembled monolayer was used to biofunctionalise the nanoparticles. We showed in Ref. [94] that the binding of the single stranded capture oligonucleotides by thiol-gold interaction and the hybridisation of the complementary DNA can be detected on single nanoparticle level, where each binding step leads to a spectral shift of LSPR. Particularly, different immobilisation strategies were examined for their efficiency and it was demonstrated that the molecular weight of the DNA molecules has a non-linear effect on the spectral shift of the LSPR [see Fig. 5(a)].

The sensitivity of the nanoparticles plays a key role in biomolecule detection and can be adjusted by the form and material of the nanostructures. Our group investigated the sensitivities of different core-shell nanostructures towards changes in the surrounding refractive index [95]. In Fig. 5 the sensitivities of the core-shell nanoparticles are compared to gold and silver spherical nanoparticles and gold nanorods. Certain core-shell nanoparticles show significant higher sensitivities compared to other nanoparticle types. The shell thickness plays an important role, e.g. the core-shell nanoparticles with the thickest silver shell show comparable results to pure silver nanoparticles. The effect of enhanced sensitivity was explained by the existence of two overlapping plasmon bands in the NP, which change their relative intensity upon change of refractive index. This indicates that producing an extra designed metallic shell around simple spherical NP increases their usability as a transducer in LSPR sensing.

In a standard configuration, a detection of LSPR spectra of single nanoparticles has to be carried out in the dark-field configuration. It means that the illumination light has to fall on the probe under high angles so that it is not reflected to the collection optics. This method of background signal suppression can be technically challenging, therefore, we used a different approach [96, 97]. A directly illuminated non-transparent chromium film with sub-wavelength apertures was used instead. These holes transmitted only a fractional part of the light, which was recorded by a detector. If a nanoparticle was present in the nanohole, the intensity of the transmitted light increased and the spectral transmission characteristic changed. The increased transition was for the wavelength above the plasmon resonance of the nanoparticle, which also a numerical simulation confirmed. It was possible to simultaneously identify a number of nanoparticles in all holes by analyzing only the intensity of light in holes recorded by a RGB camera [see Fig. 5(c)]. DNA hybridization experiments on single nanoparticle in the nanohole were also carried out. By using micro-spectroscopy the spectra were recorded for each immobilization and hybridization step. The results showed that each step lead to a detectable change of plasmon resonance [see Fig. 5(d)] suggesting that this method can be an alternative to dark-field illumination set-up.

2.3.2 Electric detection of small molecules by nanoparticles

Besides exploiting the optical properties of metallic nanoparticles for detection, also their electric properties can be used. The following method was developed in our group to detect DNA probes [98–101]. The detection chips consist of micro-structured electrodes array, where single stranded DNAs complementary to the detecting DNAs are attached in the gaps between them. After the sample solution is brought over the electrodes, the complementary DNAs attached with gold nanoparticles hybridizes with DNA in the gaps. In this step there is still no electrical connection established between the electrodes, because the nanoparticles are not touching each other. Therefore, a silver enhancement solution is used to create silver shell around the nanoparticles. If sufficient time for the reaction is allowed, the nanoparticles grow together and form a conducting layer (see Fig. 6 left), which bridges the electrodes and decreases the resistivity between the electrodes. We showed that it is also possible to use enzymes instead of initial gold particles, which also promote the growth of silver nanoparticles [101]. Because of the simple electric read-out, this detection method can be easily integrated into portable lab-on-a-chip device and detection plants pathogens. This was demonstrated by building a prototype for plants pathogen detection [100].

Another way of electrical detection of small molecules is measuring the resistivity of nanoparticle bridges created between electrodes and assembled by Dielectrophoresis [76]. These bridges exhibit low resistivity, because the gold nanoparticles are touching each other. If a solution with molecules preferably attaching to the NP is placed over the electrodes, the bound molecules influence the charge transfer (increase the resistivity) along the particle chain. By bridges with appropriate geometry, it is possible to monitor on-line the adsorption of molecules to the particles as seen in Fig. 6.

2.4 Controlled energy guiding with nanostructures

The metallic nanoparticles are excellent light absorber at their plasmon resonance wavelength. This effect can be used to concentrate energy into the small volume of the nanoparticles. In Ref. [102] we theoretically and experimentally investigated the dependence of the laser pulse duration on energy absorption to the nanoparticle and consequent energy transfer from nanoparticles to the surrounding medium. The energy transfer from nanoparticle to the medium was monitored by mapping morphological changes of a substrate, where the nanoparticle was positioned. It was shown that if femto-second laser pulses with two photons adsorption are utilized, materials only in the vicinity of the particles were destroyed. Therefore, this method can be applied for multi parallel cutting of DNA strands [103] or in sub-wavelength photo lithographic procedures.

Extension of this plasmonic nanolithography was shown in Ref. [104]. In this work, metal nanoparticles with micrometers long DNA strands were deposited on polymer substrates. When the NPs are illuminated with femtosecond laser pulse and NPs are not in a contact with the DNA bundles then only polymer substrate destruction around the NPs is observed. But if a NP is attached to the DNA bundle, then the energy accumulated in the NPs can be coupled into the DNA strands and the energy is guided in it. This energy transport along the DNA causes polymer substrate destruction at the place of the DNA bundle. The schematics of the effects are together with AFM images of the NP labeled

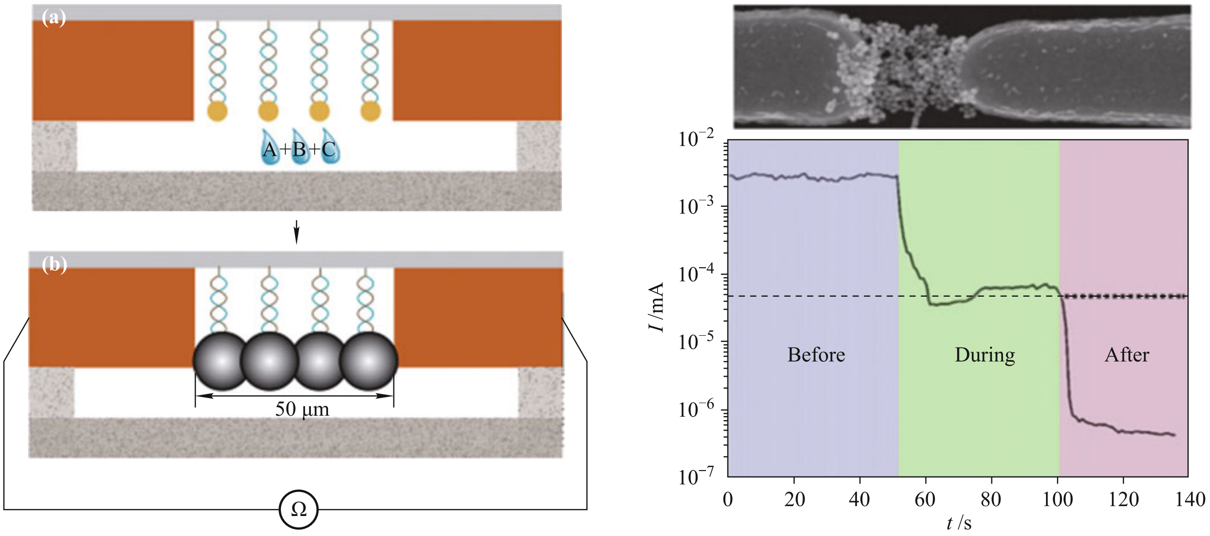

Fig. 6 *Left*: After immobilization of single stranded capture-DNA sequences, the nanoparticle or enzyme labelled target-DNA hybridizes to its specific partner on the DNA chip **(a)**. In the next step **(b)**, silver enhancement reaction connect the electrodes. *Right*: Real-time resistance measurements when the analyte molecules are applied thus monitoring the binding kinetics of thiol modified molecules. Reproduced from Refs. [101, 76] with permission from Elsevier.

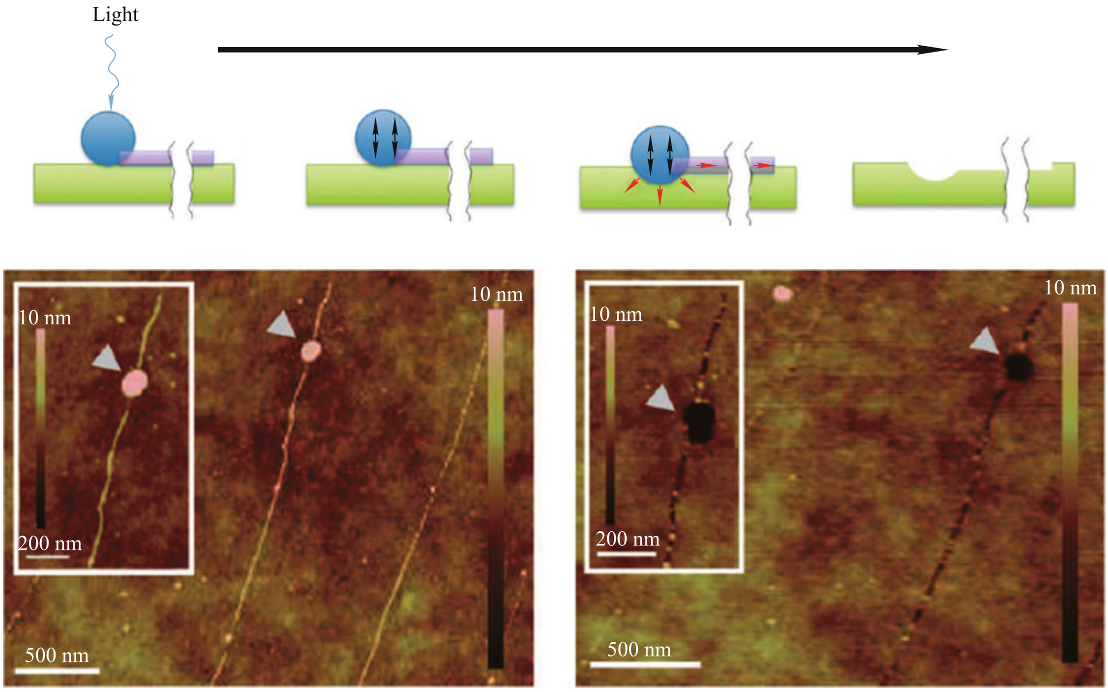

Fig. 7 *Top*: A diagram of nanowire-based plasmonic lithography, where the light induced excitation of metal nanostructures is guided along molecular nanowires. *Bottom*: AFM images of nanoparticle-labeled DNA bundle structures before (*left*) and after (*right*) pulsed laser irradiation. Adapted with permission from Ref. [104], Copyright © 2011 American Chemical Society.

DNA bundles before and after pulsed laser irradiation in Fig. 7. The AFM images show that the energy can be guided as long as several micrometers away from the metal nanoparticles, but still highly localized on the DNA bundle. Because the DNA molecules can be folded in a highly controlled manner [105], this method opens new ways in optical lithography with nanometer high resolutions. The energy transfer along the DNA bundle was also independently confirmed by a photo bleaching of fluorescent molecules attached to the DNA bundles [106]. Also the energy can by guided without destruction of the DNA bundle. This can be exploited to build interconnection in highly integrated molecular devices.

3 Summary

In this review, the concept of plasmonics was introduced and its latest development and applications were highlighted. The overview of different nanostructures supporting plasmon resonances in zero, one and two dimensions were presented. Directions of the research in the field of plasmonic, which were carried out by our group, were highlighted. It was shown that bi-metallic core shell nanoparticles have interesting plasmonic properties and their production can be improved by the concept of microfluidic segmented flow. This concept can be expanded to the production of other types of nanoparticles. For synthesis of conjugated nanoparticle complexes the construction based on DNA is applied. It was shown that nano-objects can be precisely manipulated and incorporated into a designated area, which aims to overcome the limitation of "bottom-up" synthesis. Afterwards, several sensing principles with plasmonic nanostructures were demonstrated. The LSPR sensing on a single NP or in a combination with a sub-wavelength aperture were investigated in detail. It was shown that also the electrical properties of NP assemblies can be utilized to build robust detectors for bio-molecules sensing. Finally, we presented the results with energy focusing and guiding with nanometer resolution, which can be in future used for an ultra-precise optical lithography or as connections in molecular devices.

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