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Plasmonic biosensors for food control

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ABSTRACT

Background: Food safety is becoming increasingly important because food industry must provide quality products to minimize the health risks. Traditional methods to assure food safety, such as plate count and polymerase chain reaction are accurate and robust but can hardly satisfy the needs of the food industry because they are costly and time consuming. Therefore, optical biosensors that can analyze food in a low-cost, facile, fast, sensitive, and selective manner started to emerge.

Scope and approach: This review presents plasmonic biosensors including surface plasmon resonance (SPR), localized SPR (LSPR), fiber optic SPR (FO-SPR), surface enhanced Raman scattering (SERS), surface-enhanced fluorescence (SEF), and total internal reflection (TIR) based sensors and their applications in food pathogens monitoring. Moreover, the strengths and weaknesses of plasmonic biosensors implementation in food control are showcased.

Key findings and conclusions: Plasmonic biosensors could simplify procedure and radically reduce time, price and consumption of reactants, compared to traditional microbiological methods. Optical biosensors, in particular SPR, have been developed for detection of different foodborne pathogens. In parallel, analytical improvements have been achieved by coupling different techniques (fiber optics, Raman, fluorescence, luminescence) to plasmonic sensors in order to reduce the limits of detection and to improve sensitivity. The future improvements include the miniaturization of instruments to handheld devices and simplification of analysis to enable direct target detection in food matrices. Plasmonic technology can certainly have long lasting impact because the need for a simple and rapid food assay is pressing and guarantees the future development in this field.

1. Introduction

Food control has an increasing importance for food industries due to globalization and climate changes. Various foods are delivered all over the world, allowing consumers to taste different kinds of foods but also bringing potential risks for food safety. The complexity of most foods makes them difficult to be analyzed with conventional microbiological techniques. Currently, the plate count methods and polymerase chain reaction (PCR) are mostly used for microbial control, while gas chromatography and high pressure liquid chromatography are used for chemical control of food. All these methods have some issues such as the requirement of trained personnel, cost and time consumption. Plate

count methods take up to 7 days to provide results (from enrichment step to strain identification test), which is a too long time for foods with a short shelf-life. PCR methods reduce the assay time to 24 h–48 h, but can be sensitive to complex food matrices compounds, which can inhibit DNA polymerase and produce false negative results.

Based on European Food Safety Authority (EFSA) report, *Campylobacter*, *Salmonella*, *Yersinia*, *E. coli* and *Listeria* are the most widespread food pathogens in the European Union. They are responsible for over 350,000 human cases each year, but the estimated real number is much higher (Authority, Prevention, & Control, 2018; Lachenmeier, Löbell-Behrends, Böse, & Marx, 2013). To protect population, an accurate survey from raw materials to the final product including the

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processes used for food stabilization, transportation and distribution is needed (Thakur & Ragavan, 2013). Fully integrated, handheld, sensitive and inexpensive point-of-care devices able to provide results in a short time are under high demand by food industry to enable rapid analyses along the whole food chain, in on-line controls, and at market level. The market of the analytical tests developed for food safety applications, especially for pathogen detection, was estimated around \$4 billion in 2018 in Europe, a value which proves the economic importance of the sector (Vidic et al., 2019).

Optical biosensors are among the most studied devices due their relatively low cost and easy utilization (Kim, Lee, Lee, Song, & Gu, 2019). They are composed of a biosensing layer (containing antibodies, aptamers, DNA probe, or other biological molecules) aiming to recognize the target, and an optical transducer that converts the recognition event into a measurable signal. Recent developments in plasmonic sensors (Fig. 1), that are based on an optical phenomenon generated by light when it interacts with conducting interfaces in thin film form and nanoparticles that have smaller sizes than the incident wavelength, led to fabrication of biosensors with a high sensitivity and selectivity. Plasmonic biosensors that can be applied to solid and liquid food analyses require few μL sample and may be easily miniaturized. The aim of this review is to highlight the effectiveness of plasmonic biosensors for detecting food pathogens present in contaminated samples.

2. Optical and plasmonics biosensors

Optical biosensors can be divided in labeled and label-free. Labeled techniques are expensive and complex, as they require several steps to be performed. DNA biosensors based on organic light emitting diode (OLED) use a couple of DNA probes: one probe, called capture probe, is immobilized onto the glassy transducer and a secondary probe, called detection probe, is labeled with a fluorescent tag or with an enzyme to produce light and allow revelation of the target (Manzano et al., 2015). Also optical fibers based DNA biosensors may utilize two DNA probes, the capture probe is immobilized onto the glassy core of the fiber, and a secondary (detection) labeled probe is used to reveal the target (Cecchini, Manzano, Mandabi, Perelman, & Marks, 2012). In contrast,

label-free biosensors can directly detect interaction of the target with the capture probe immobilized on the transducer layer.

The label-free biosensing is the most commonly used so far in a wide range of applications because it provides fast response, simple utilization and may be self-administrated. Several optical sensors have provided a great potential to the research community to develop label-free and sensitive sensors. Surface plasmon resonance (SPR), localized SPR (LSPR), fiber optic SPR (FO-SPR), surface enhanced Raman scattering (SERS), surface-enhanced fluorescence (SEF), total internal reflection (TIR) based, interference based, waveguide based, and specifically structured evanescent field sensors, are all optical techniques which utilize non labeled DNA probes. Among these, plasmonic based sensors have been broadly used for food safety and quality analysis due to high sensitive and rapid detection of analytes, applicability of miniaturization to a handheld format, simultaneous detection of different targets in real-time and in a label free environment (Y. Chen, Liu, Yang, Wilkinson, & Zhou, 2019).

The techniques based on plasmonic sensors are SPR, LSPR, FO-SPR, SERS, SEF and SEIRA. These sensors are usually based on noble metallic thin films and nanostructures, because of their property of higher optical absorption band in the visible-near infrared range of the electromagnetic spectrum, which is called plasmonic band. Among eight noble metals, gold (Au), silver (Ag), platinum (Pt), and palladium (Pd) are preferably used in plasmonic sensors. These metals are highly conducting, reflecting, biocompatible, inert to corrosion and oxidation, features providing stability. Silver, which has excellent plasmonic properties but oxidizes relatively fast, can be covered with a thin protection layer such as ZrO_2 or SiO_2 . SPR is the phenomenon attributed to the resonant coupling of the evanescent field of transverse magnetic (TM) polarized light with an oscillating free electron wave bound at the metallic-dielectric interface. At resonance condition, the incident TM polarized wave will show an attenuation while collecting the reflected light at a specific resonance parameter, depending upon the mode of interrogation (angular or spectral interrogations, usually) employed. Fig. 2A represents a conventional prism based Kretschmann configuration to achieve SPR, which is in general used to develop SPR based biosensors (Kretschmann & Raether, 1968). The depth and position of SPR dip is sensitive to the

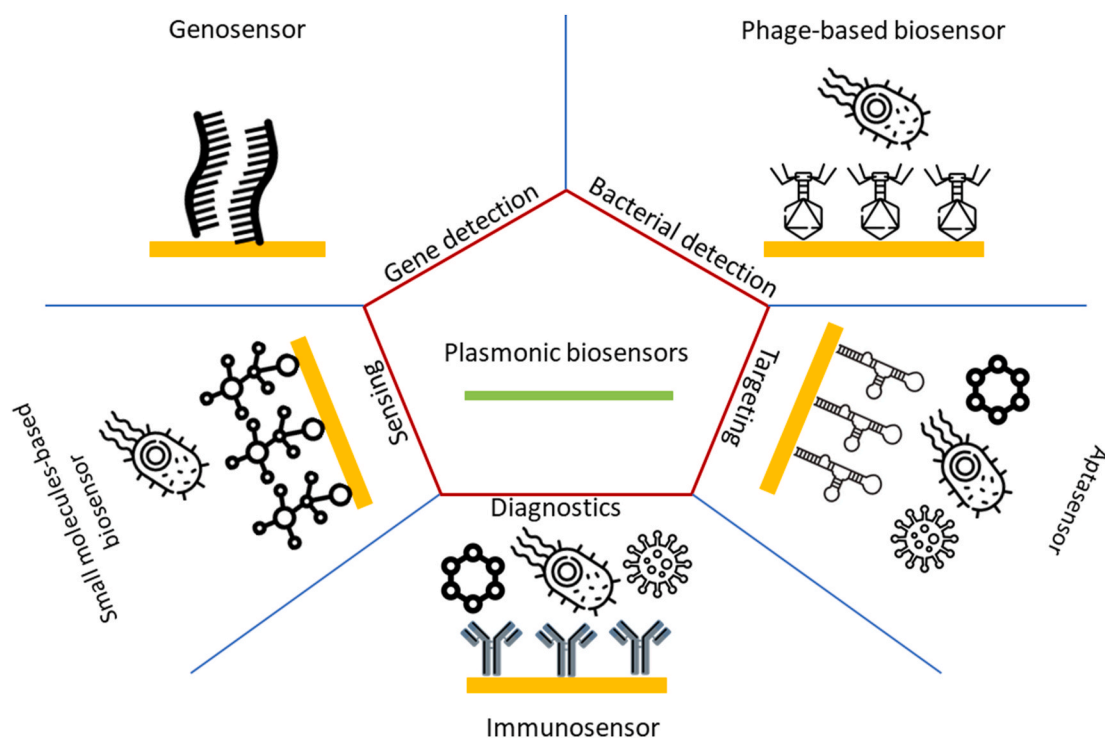


Fig. 1. Schematic illustration summarising examples of the more commonly used recognition elements in various branches of plasmonic biosensors.

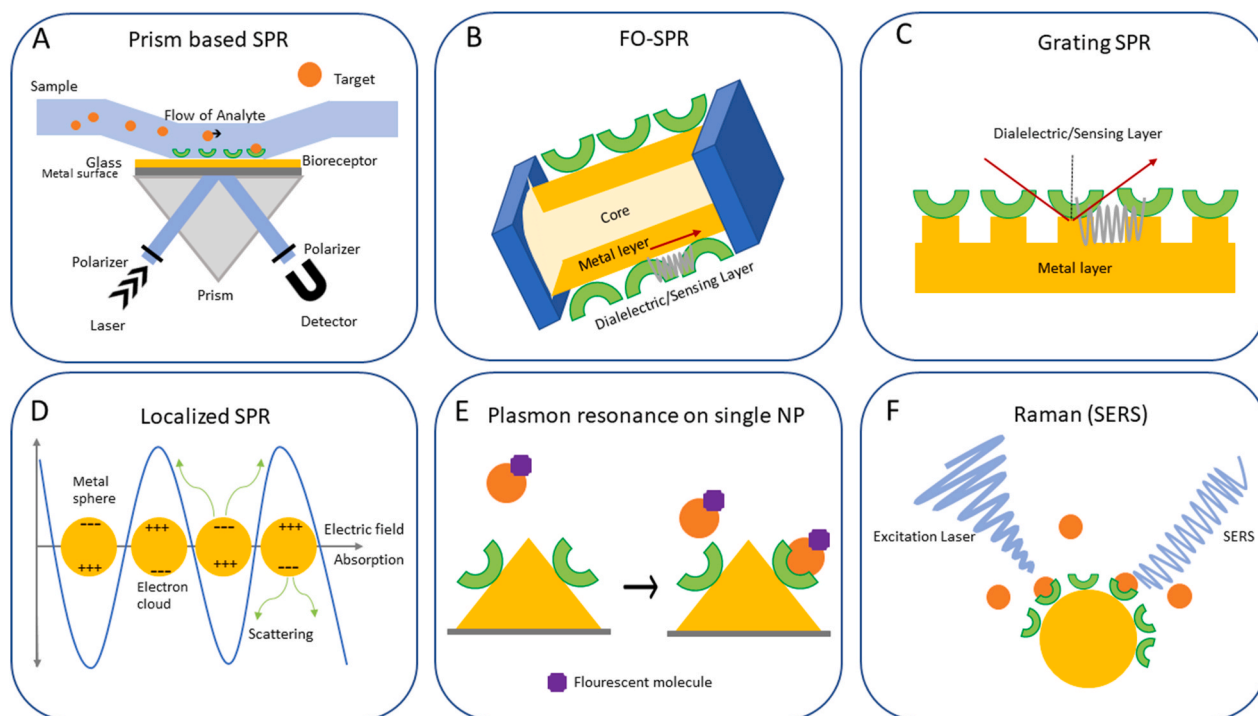


Fig. 2. Plasmonic biosensors fundamentals. Various configurations to realize SPR sensor: prism based (A), optical fiber based (B) and grating structure (C). Pictorial representation of electromagnetic field with metallic nanoparticle in LSPR (D). Schematic representation of SEF (E) and SERS (F).

refractive index of the overlayer, based on which, the SPR sensors are developed for a quantitative target detection and a range of applications.

Gold and silver chips are mostly used in SPR biosensors, because they can be easily functionalized with the biomolecules and exhibit higher sensitivity comparable to other plasmonic materials such as aluminum and copper (B. D. Gupta, Shrivastav, & Usha, 2017). In spite of this, various groups have used other metals or their combinations to enhance the performance of SPR sensors such as bimetallic SPR (B. Gupta & Sharma, 2005), long range SPR (Isaacs & Abdulhalim, 2015), guided wave SPR (Lahav, Auslender, & Abdulhalim, 2008; C. Lin & Chen, 2019), or self-referenced plasmon waveguide SPR (Harpaz, Koh, Seet, Abdulhalim, & Tok, 2020). As an example, Isaacs and collaborators developed a self-referenced Plasmon Waveguide Resonance (PWR) sensor that consisted of an insulator-metal-insulator chip to achieve resonance using both TE and TM polarizations, resulting in a narrower resonance of classical SPR based sensor (Isaacs, Harté, D Alves, & Abdulhalim, 2019). SPR based sensors have several advantages compared to conventional sensors including label-free detection, high sensitivity, and applicability to real-time kinetic measurements. However, they suffer from some limitations such as the requirement for TM polarized light, exhibit shallow penetration depth (200–300 nm), and have low selectivity. Small penetration depth is useful for selective detection of small molecules or viruses near the vicinity of the plasmonic surface, while for large bio-entities a long range SPR, or guided wave SPR, are more appropriate such as the one developed by Isaacs and Abdulhalim (Isaacs & Abdulhalim, 2015). The optical fiber configuration used to excite SPR named fiber optic SPR (FO-SPR), as shown in Fig. 2B, has the advantages of compact size, chemical inertness, immunity of external electromagnetic interference, multi-channel and multi-analyte advances along with applicability of online monitoring and remote sensing (B. Lee, 2003; Sharma, Jha, & Gupta, 2007). However, with respect to conventional prism based SPR, FO-SPR suffers from higher broadening due to number of reflections of many rays or existence of skew rays occurring at the core-metal surface (Sharma et al., 2007). Besides, prism or optical fiber configuration, SPR can also be achieved by grating configuration, schematically showed in Fig. 2C.

In case of metallic nanostructures, the resonant field associated with surface plasmons is localized then SPR is termed as localized SPR (LSPR) (K. M. Mayer & Hafner, 2011; Roh, Chung, & Lee, 2011); where resonance wavelength depends upon the shape and size of plasmonic nanomaterial and the medium surrounding it. Based on this, LSPR sensors are realized with receptors functionalized on the plasmonic nanostructured material (Nehl & Hafner, 2008). Fig. 2D shows the schema of an electromagnetic interaction of light with metallic nanoparticles (Roh et al., 2011). Due to the use of nanostructures instead of semi-infinite plasmonic surfaces, LSPR possesses the high surface-to-volume ratio for sensing surface-analyte interactions, which can be integrated into more compact and miniaturized devices for higher applicability. The main disadvantage of LSPR is its lower refractive index sensitivity compared to conventional SPR (Cao, Galbraith, Sun, & Grattan, 2011). Information on SPR and LSPR can further be found in several reviews (Abdulhalim, 2013; Esfahani Monfared, 2020; Roh et al., 2011; Shalabney & Abdulhalim, 2011; Jingwei; Sun et al., 2020; Szunerits, Shalabney, Boukherroub, & Abdulhalim, 2012). Plasmonic properties of a single nanoparticle can be analyzed and monitored by using dark-field microscopy (Anker et al., 2010; Sriram et al., 2018; Sriram, Zong, Vivekchand, & Gooding, 2015). The color analysis of scattered light from plasmonic nanoparticles, which is directly related to the LSPR of the nanoparticle, its size, shape, and environment, allows to detect single analyte-ligand binding events.

Surface enhanced spectroscopies are also domains related to plasmonics, which improve the performance of various spectroscopic techniques such as Raman, fluorescence or infrared absorption to measure the qualitative and quantitative response of the sensor. In surface enhanced fluorescence (SEF), the metallic nanoparticle is placed near the proximity of the fluorophore so that the localized plasmonic field due to LSPR coupled with fluorophore electrons results in the enhanced fluorophore intensity (Bauch, Toma, Toma, Zhang, & Dostalek, 2014; Fothergill, Joyce, & Xie, 2018), as schematically shown in Fig. 2E. SEF is used in a number of applications such as specific sensing, molecular characterization, cellular imaging, and single molecule spectroscopy. Energy quenching sometimes limits the performance of the devices

based on SEF, which can be reduced by selecting appropriate parameters such as the fluorescence material (quantum dots instead of organic molecules), nanostructures, emission intensity, radiative decay, etc. Similarly, to achieve SERS, the Raman active molecule is placed within the localized electric field of the plasmonic nanostructures such that the molecule will experience an increased Raman scattering cross-section and hence, enhanced Raman signal (Barbillon, 2020; Hamm, Gee, & De Silva Indrasekara, 2019; P. Li et al., 2020; Shvalya, Filipič, Zavašnik, Abdulhalim, & Cvelbar, 2020). Due to the specific fingerprints of fluorescence at molecular level, SERS has shown several advantages of high selectivity, single molecule detection, and molecular characterization, and in addition high throughput, point-of-care applicability, and easy sample preparation. However, it suffers from the requirements of sophisticated instrumentation to provide high reproducibility for the intensity measurement. Fig. 2F represents a pictorial representation of a configuration to achieve SERS. The enhancement of IR signal is obtained by placing the material near the plasmonic nanoparticle. Moreover, due to IR wavelength activity SEIRA can be achieved by a wide range of plasmonic materials including semiconductors, graphene, and metals (Brehm, Taubner, Hillenbrand, & Keilmann, 2006; Yang et al., 2018). This technique suffers from lower enhancement factor (10–1000) compared to SERS and SEF, where enhancement factors up to 10^6 – 10^8 orders are reported.

The simulations and design of plasmonic biosensors is also one of the most important aspects to be considered before its fabrication. Sensor performance depends upon number of parameters including the plasmonic material, the dielectric properties, associated electromagnetic field, and properties of incident light. The simulations for the SPR based plasmonic sensors are usually performed by estimating the reflectivity and corresponding electromagnetic field at each layer by well-known *N*-layer model, which is mainly based on 2×2 Abeles matrix approach (Born & Wolf, 2013; Shalabney & Abdulhalim, 2011). In case of FO-SPR, similar method is used just by integrating the multiple reflections from critical angle (with respect to metal-core interface) to 90° (Sharma et al., 2007). For LSPR design, Mie or discrete dipole approximation scattering based theories are used for single particles while for more complicated nanostructures finite domain time domain (FDTD) or finite element (FEM) numerical methods are used with powerful software packages such as COMSOL and Numerical.

Plasmonic biosensors are powerful tools for contaminant detection in food samples (Table 1). Their excellent analytical performances reflect the sensitivity of transducers that can provide recognition signal directly in complex food matrices. Bacterial biomarkers or food contaminants can be directly detected in liquid foods like in milk, wine or fruit juice using standard SPR biosensors (Jiang, Sun, Pu, & Wei, 2019; Mihai et al., 2015; Pilolli, Visconti, & Monaci, 2015; Todescato et al., 2014; Yi, Wang, Dostálek, & Knoll, 2009; Zhao, Feng, Hu, Wang, & Lu, 2019; Z.; Zhu et al., 2015). The analysis of solid foods can be also performed by SERS/SEF/LSPR techniques by placing the sample on top of plasmonic substrate or by adding nanoparticles on top of the solid food (Fu, Sun, Pu, & Wei, 2019; B.; Liu et al., 2012; Oh et al., 2017; Sabet, Hosseini, Khabbaz, Dadmehr, & Ganjali, 2017; Tang et al., 2012; S.; Wang, Dong, & Liang, 2018).

2.1. Plasmonic immunosensors

In immunosensors, the formation of antibody-antigen complex is converted to a signal that can be processed and recorded, as illustrated in Fig. 3. Immuno-sensor-chip can be easily re-used after its regeneration by dissociating the immobilized antigen from the surface. Limitations in immunosensors include the antibody' high cost, limited lifespan and susceptibility to biodegradation (Thakur & Ragavan, 2013; Vidic, Manzano, Chang, & Jaffrezic-Renault, 2017). However, thermal stability, solubility and strength against environmental stress can be improved by antibody tailoring or by using only its heavy chain or a small domain bearing the binding site, called nanobodies (Hwang, Korves, Renda, &

Suh, 2012; Lim & Ahmed, 2016).

A direct immunoassay for bacterial detection provides a limit of detection (LOD) of about 10^6 CFU/mL (J. Chen & Park, 2018; Masdor, Altintas, & Tothill, 2017), which is usually highly above the minimum infectious doses. To improve sensitivity, immunoassays are performed after an enrichment step (J. Chen & Park, 2018; X. Zhang et al., 2017) or in a format which enhances signal-to-noise ratio. For instance, *Campylobacter jejuni* was detected with a 2 log higher sensitivity in a sandwich amplification format compared to direct detection using SPR (Masdor et al., 2017). The SPR instrument equipped with multiple sensing spots enables to perform separate assays simultaneously. One spot was functionalized with the specific *anti-C. jejuni* antibody (active channel) while another spot was functionalized with mouse IgG (control channel). In direct assay, *C. jejuni* cells were injected over two spots to provide resulting SPR signal proportional to the amount of bound cells. In sandwich assay, after capturing *C. jejuni* cells on the active channel, the specific antibody was injected to further increase the signal (Fig. 4A). Finally, the replacement of the antibody by antibody-conjugated gold nanoparticles (Au NPs) additionally increase the detection signal. A LOD of only 130 CFU/mL was obtained in a subtractive inhibition assay in which the rabbit polyclonal antibody specific for *C. jejuni* was first mixed with bacterial cells, and then unbound antibody was separated and detected using an immobilized anti-rabbit IgG on the SPR chip (Fig. 4B) (Masdor, Altintas, Shukor, & Tothill, 2019). Alternatively, magnetic nanoparticles decorated with a specific antibody enable enhancement of SPR signal read-out (X. Liu et al., 2016; Yuling Wang, Ravindranath, & Irudayaraj, 2011). A LOD of only 14 CFU/mL was reached when magnetic nanoparticles directed against *Salmonella enteritidis* were used as both a bioreceptor and an amplification reagent (X. Liu et al., 2016). Furthermore, a LOD as low as 3 CFU/mL was obtained for detection of *Escherichia coli* in a method combining the SPR spectroscopy and immuno-magnetic separation using gold-coated magnetic nanoparticles (Torun, Boyaci, Temür, & Tamer, 2012).

Utilization of a multichannel surface provides a platform for simultaneous immuno-detection of different bacteria. Such devices can be miniaturized to a portable format (Morlay et al., 2017; Taylor et al., 2006; X. Zhang et al., 2017). By combining a SPR multichannel biosensor and a simultaneous enrichment step, Zhang and collaborators (2017) developed an immunoassay for the detection of *E. coli* O157:H7, *S. enteritidis* and *Listeria monocytogenes* in chicken meat with LOD of 14, 6, and 28 CFU/25 g, respectively (X. Zhang et al., 2017). Specific *anti-E. coli* O157:H7, *anti-Salmonella*, and *anti-Listeria* antibodies were immobilized onto the surface of different channels in a multichannel SPR biosensor using the amine-coupling method. The immobilization consisted in activation of carboxyl groups of PEG6 molecules grafted to the gold surfaces and subsequent antibody injections. Then sensor surface was blocked with ethanolamine and albumin to prevent nonspecific interactions. Two flow channels carried no antibody served as the reference channels (Fig. 4C). Coupled with a microfluidic, SPR biosensor could capture and detect pathogens in an automatic way (Tokel et al., 2015). Such systems can potentially be used for detection of any bacterial or viral pathogen.

Similar to SPR, SERS has also been widely used to design and develop highly sensitive biosensors for food control. Detection of various food-borne pathogens such as *E. coli* O157:H7, *Staphylococcus aureus*, *Staphylococcus epidermidis*, and *Salmonella* Typhimurium was demonstrated by Chu et al. using Ag nanoparticles as SERS substrate (Chu, Huang, & Zhao, 2008). The biosensor successfully discriminated the bacteria in terms of Gram-stain types, species, strains, and viability. Liu et al. reported Au@Ag core-shell nanoparticles as SERS platform to detect and identify the pesticide residues (such as thiocarbamate and organophosphorous compounds) over various fruit peels (B. Liu et al., 2012). This study showed that the SERS response of the pesticides is two-fold enhanced compared to bare Ag or Au nanoparticles. The detection limits of pesticide sensors were found within 1.5 ng/cm^2 over the peel

Table 1
Plasmonic biosensors application in food analysis.

arget	Optical method	Substrate	Limit of detection	Recognition element	Reference
<i>C. jejuni</i>	SPR (direct, sandwich and sandwich with AuNPs amplification)	Gold	4 × 10 ⁴ CFU/mL (sandwich) 8 × 10 ⁶ CFU/mL (direct) 8 × 10 ⁵ CFU/mL (sandwich with AuNPs amplification)	Antibody	Masdor et al. (2017)
<i>S. enteritidis</i>	SPR (sandwich immunoassay)	Fe ₃ O ₄	14 CFU/mL	Antibody	(X. Liu et al., 2016)
<i>S. enteritidis</i> , <i>S. Typhimurium</i> , <i>S. heidelberg</i>	SPR	Gold	2.1 × 10 ⁶ CFU/ml in PBS; 7.6 × 10 ⁶ CFU/mL in chicken rinse matrix, 6.8 CFU/mL after enrichment step		(J. Chen & Park, 2018)
<i>Campylobacter</i> spp.	SPR (subtractive inhibition assay)		131 ± 4 CFU/mL	Antibody	Masdor et al. (2019)
<i>C. jejuni</i>	SPR	Gold	10 ³ CFU/mL	Antibody	Wei et al. (2007)
<i>E. coli</i> O157:H7, <i>S. choleraesuis</i> serotype Typhimurium, <i>L. monocytogenes</i> , <i>C. jejuni</i>	Eight channel SPR	Gold	(3.4 × 10 ³ - 1.2 × 10 ⁵) CFU/mL in buffer and apple juice (pH 3.7 and 7.4)	Antibody	Taylor et al. (2006)
<i>E. coli</i> O157:H7, <i>S. enteritidis</i> and <i>L. monocytogenes</i>	SPR	Gold	14 CFU/25 g (<i>E. coli</i>); 6 CFU/25 g (<i>S. enteritidis</i>), 28 CFU/25 g (<i>L. Monocytogenes</i>)	Antibody	(X. Zhang et al., 2017)
<i>E. coli</i>	SPR (subtractive inhibition assay)	SPR	3.0 × 10 ⁴ CFU/mL	Antibody	(Yixian Wang, Ye, Si, & Ying, 2011)
<i>E. coli</i>	SPR	Gold	3 CFU/mL	Antibody	Torun et al. (2012)
<i>E. coli</i>	SPR (sandwich assay)	Gold	10 ⁴ CFU/mL	Antibody	Tokel et al. (2015)
<i>L. monocytogenes</i>	SPR imaging	Gold		Antibody	Morlay et al. (2017)
Porcine circovirus 2 (PCV-2)	LSPR, SPR	Gold		Antibody	Basso et al. (2020)
Pesticides	SPR	Gold	3.5 ng/mL (azoxystrobin), 4.5 ng/mL (boscalid), 2.5 ng/mL (chlorfenapyr), 5.5 ng/mL (imazalil), 3.5 ng/mL (isoxathion), 8.5 ng/mL (nitenpyram)	Antibody	Miyake et al. (2019)
Ochratoxin A	SPR (colloidal gold particles)	Gold	0.06 ng/mL	Antibody	Urusov, Kostenko, Sveshnikov, Zherdev, and Dzantiev (2011)
Aflatoxin B1	SPR	Gold	1 µg/mL	Antibody	Park et al. (2014)
Ractopamine	SPR	Gold	0.6 µg/kg in pork meat sample	Antibody	Lu et al. (2012)
Tetrodotoxin	SPR	Gold	sub-ng/mL	Antibody	Yakes, Deeds, White, and DeGrasse (2011)
Peanut allergens (Ara h1)	FO-SPR	Gold	0.09 µg/mL	Antibody	Pollet et al. (2011)
13 food allergens	Imaging SPR array		2 mg/mL	Antibody	Rebe Raz et al. (2010)
Proteins from milk, egg, hazelnut, peanut, shellfish, and sesame	SPR (direct and sandwich)		1–12.5 µg/g	Antibody	Yman et al. (2006)
α-casein	SPR	Gold	58 ng/mL	Antibody	Ashley et al. (2017)
Ovalbumin	SPR	gold	0.03 and 0.2 µg/mL	Antibody	Pilolli et al. (2015)
Aflatoxin B ₁	SERS	Silica encapsulated Au Nanoparticles	0.1 ng/mL	Antibody	Ko, Lee, and Choo (2015)
<i>L. monocytogenes</i>	SERS	AuNP@Ag	27 CFU/mL	Antibody	(H.-b. Liu, Du, Zang, Li, & Wang, 2017)
<i>S. enterica</i>	SERS	AuNP@Ag	19 CFU/mL	Antibody	(H.-b. Liu et al., 2017)
<i>E. coli</i> O157:H7, <i>S. typhimurium</i> , <i>S. aureus</i>	SERS	Ag nanospheres	10 CFU/mL	Antibody	(Yuling Wang, Ravindranath, & Irudayaraj, 2011)
<i>S. typhimurium</i> , <i>S. aureus</i>	Magnetic SERS	Fe ₃ O ₄ NPs@ SiO ₂ -Au NPs	10 ³ CFU/mL	Antibody	(Yuling Wang, Ravindranath, & Irudayaraj, 2011)
<i>E. coli</i> O157: H7, <i>S. epidermidis</i> , <i>L. monocytogenes</i> , <i>E. faecalis</i>	SERS	Ag nanoparticles	Single cell detection	Antibody	Fan, Hu, Mustapha, and Lin (2011)
<i>E. coli</i>	SERS (sandwich immunoassay)	Au nanorods	10 CFU/mL	Antibody	Temur, Boyacı, Tamer, Unsal, and Aydogan (2010)
Tricyclazole	SERS	Ag colloid	0.002 mg/l in paddy rice	Antibody	Tang et al. (2012)
Pesticides (from fruit peels)	SERS	Ag@Au core-shell nanoparticles	1.5 ng/cm ² (thiram) 0.14 µg/cm ² (chlorpyrifos) 0.1 µg/cm ² (methyl parathion)	Antibody	(B. Liu et al., 2012)
melamine	SERS	Au coated Si wafer	0.1% (in wheat gluten) 0.05% (in chicken feed) 0.05% (in cake) 0.7% (in noodle)	Antibody	(M. Lin et al., 2008)
sudan, thiram, thiabendazole	SERS (Microdroplet captured tape)	Au nano-dendrites; R6G as Raman indicator	10 ⁻⁵ M	Antibody	He et al. (2020)
thiabendazole	SERS	Au nanorods array	0.06 ppm in apple	Antibody	Fu et al. (2019)
Aflatoxin M ₁	SERS		0.06 pg/mL	Antibody	(Yi Wang et al., 2009)

(continued on next page)

Table 1 (continued)

target	Optical method	Substrate	Limit of detection	Recognition element	Reference
	Long range enhanced fluorescence	Anti-AFM ₁ functionalized Au thin film			
Ochratoxin A	Long range enhanced fluorescence	Ag-FON (Ag film over nanosphere)	0.5 µg/kg in wheat milk and apple juice	Antibody	Todescato et al. (2014)
<i>P. aeruginosa</i>	SEF	Ag nanoparticles	1.5 CFU/mL	Antibody	Ellairaja et al. (2017)
<i>E. coli</i> O157:H7	Long range surface enhanced fluorescence	Cytop/Au/antibody	10 CFU/mL	Antibody	(Yuling Wang, Ravindranath, & Irudayaraj, 2011)
<i>E. coli</i> O157:H7	Metal enhanced fluorescence	Au@Ag nanorods	3.3×10^{-18} M	Antibody	(Jiadi Sun et al., 2015)
<i>C. jejuni</i>	SPR	Gold	2.5 nM of DNA	DNA probe	Gnanaprakasa et al. (2011)
<i>S. enterica</i> serovars typhi	SPR	Gold	0.019 ng/mL	DNA probe	(Anu Singh et al., 2015)
<i>Salmonella</i> spp.	SPR	Gold	10 ² CFU/mL	DNA probe	(D. Zhang et al., 2012)
<i>Brucella melitensis</i>	SPR	Gold	<10 nM	DNA probe	Sikarwar et al. (2017)
<i>C. jejuni</i> , <i>C. coli</i>		Gold nanorods	10 ² copy/mL of amplicons	DNA probe	Shams et al. (2019)
<i>Brettanomyces bruxellensis</i>	LSPR	Gold	0.1 ng/mL	DNA probe	Manzano et al. (2016)
GMOs	SPR		20 pM (Roundup Ready) and 16 pM (lectin)	DNA probe	Plácido et al. (2020)
<i>S. enteritidis</i>	SPR	Gold	2 CFU/mL	Aptamer	Di et al. (2017)
<i>L. monocytogenes</i>	SPR (aptamer based sandwich)	Gold	20 CFU/mL	Aptamer	(S.-H. Lee et al., 2015)
<i>L. monocytogenes</i>	SPR	Gold nanorods	50 CFU/mL	Aptamer	(Y. Liu et al., 2019)
<i>S. aureus</i>	SPR (direct and bead-amplification)	Gold	1 cell	Aptamer	Chang et al. (2013)
<i>S. Typhimurium</i>	LSPR	Gold	10 ⁴ CFU/mL	Aptamer	Oh et al. (2017)
<i>P. aeruginosa</i>	LSPR	Gold	1 cell	Aptamer	Hu et al. (2018)
Allergen (Ara h1)	FO-SPR	Gold	75 nM	Aptamer	Tran et al. (2013)
Allergen (lysozyme)	SPR	Gold	0.035 µg/mL	Aptamer	Mihai et al. (2015)
Ochratoxin A	SPR	Gold	0.005 ng/mL	Aptamer	(Z. Zhu et al., 2015)
Tetracycline	SPR	Gold	0.0069 µg/kg	Aptamer	(S. Wang et al., 2018)
Bisphenol A	FO-SPR, LSPR	Germanium, Silicon dioxide, Gold	10 ⁻¹⁸ M	Aptamer	Allsop et al. (2019)
Lead (Pb ²⁺)	Resonance Scattering spectral probe	NanoGold	0.03 nmol/L	Aptamer	Ling et al. (2010)
Avian Influenza Virus (H5N2)	GO-FRET, CLSM, CD, SPR	Gold	~10 ⁵ EID ₅₀ /mL in the buffer and ~10 ⁶ EID ₅₀ /mL in the duck's feces	Aptamer	Kim et al. (2019)
Kanamycine	SERS	Double strand DNA binding Au@Ag nanoparticles	0.9 pg/mL in milk	Aptamer	Jiang et al. (2019)
Chloramphénicol	SERS	DNA modified Au Nanoparticles	15 fM	Aptamer	Fang et al. (2019)
Fumonisin B ₁	SEF	Au nanoparticles	1.1 ng/mL	Aptamer	Peltomaa et al. (2018)
Tetracycline	SEF	Ag nanoparticles encapsulated halloysite nanolumen	12.4 nM	Aptamer	Xu, Zhang, Jia, Bi, and Zhao (2020)
Aflatoxin B ₁	SEF	QDs absorbed to Au Nps	3.4 nM in peanut	Aptamer	Sabet et al. (2017)
Aflatoxin B ₁	SEF	Graphen/Au nanocomposite	0.03 pg/mL in peanut	Aptamer	(Z. Li, Xue, Ma, Cheng, & Miao, 2018)
<i>E. coli</i>	SPR		3 CFU/mL (in spinach)	Oligosaccharides	Yazgan, Noah, Toure, Zhang, and Sadik (2014)
<i>E. coli</i> O157:H7	FO-SPR	Ag nanoparticles-reduced graphene oxide	5.0×10^2 CFU/mL	Antimicrobial peptid	Zhou et al. (2018)
<i>E. coli</i> O157:H7 and methicillin-resistant <i>S. aureus</i>	SPR	Titanium and Gold	10 ³ CFU/mL	Bacteriophages	Tawil et al. (2012)
<i>Salmonella</i> spp.	SPR	Gold	3 CFU/25 g with pre-enrichment step	Bacteriophage	Karoonuthaisiri et al. (2014)
<i>B. cereus</i>	SPR (subtractive inhibition assay)	Gold	10 ² CFU/mL	Bacteriophage domain	Kong et al. (2015)
<i>E. coli</i>	SERS	Silver	1 cell/mL	Bacteriophage	Srivastava et al. (2015)
<i>C. jejuni</i>	SPR	Gold	10 ² CFU/mL	Bacteriophage	(Amit Singh, Arutyunov, McDermott, Szymanski, & Evoy, 2011)
Atrazine	Colorimetric (LSPR)	Au nanoparticles	1.1 mg/mL	Molecular imprint polymer	Zhao et al. (2019)
Atrazine	SERS	Au nanoparticles	0.0015 mg/mL	Molecular imprint polymer	Zhao et al. (2019)
<i>E. coli</i> O157:H7	SERS	Au nanoparticles	10 ² CFU/mL in Beef	Aptamer	Díaz-Amaya et al. (2019)
<i>E. coli</i> , <i>S. aureus</i> , <i>S. epidermidis</i> , <i>S. typhimurium</i>	SERS (direct)	Ag nanorods		Gold	Chu et al. (2008)

(continued on next page)

Table 1 (continued)

target	Optical method	Substrate	Limit of detection	Recognition element	Reference
Melamine	SERS	Raman active Au nanoparticles	100–200 µg/l	Gold	Mecker et al. (2012)
Mycotoxin	Surface enhanced fluorescence	Metal-organic framework	0.135 M	organic framework-derived composite	Tian et al. (2018)
Sulfamethazine	Surface enhanced fluorescence	Ag nanoclusters	0.5 µg/l	Antibody	(N. Zhu et al., 2019)

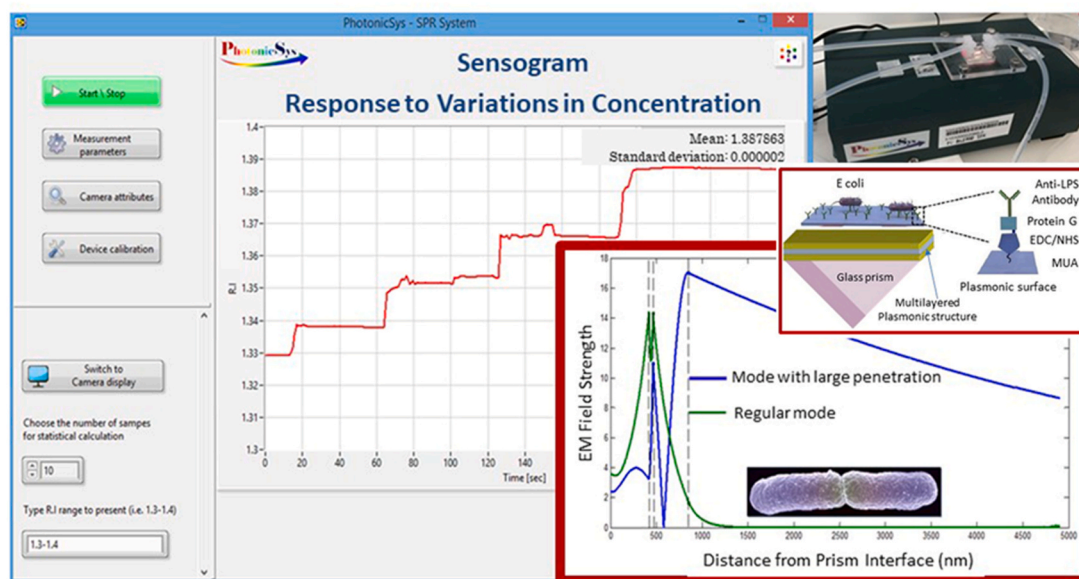


Fig. 3. An example of potential miniature handheld SPR instrument of Photonicsys Ltd., showing its user friendly interface, continuous monitoring of refractive index variations with 10^{-6} precision. The bottom right figure shows the electromagnetic field distribution for a multilayered plasmonic structure which enables tuning the penetration depth from 300 nm till few microns, hence its suitability for small and large bioentities detection. The existence of two resonances as seen provides a self-reference operation. Figures adapted with permission from Photonicsys website (www.photonicsys.com).

surface. In 2009, a combination of long range surface plasmons and fluorescence was employed by Wang et al. (2009) as SEF for the highly sensitive detection of aflatoxin M₁ in milk (Yi Wang et al., 2009). The sensor was based on inhibition assay where AFM₁ molecule derivative was immobilized over the sensor surface and corresponding antibodies were used as recognition elements. The detection limit of 0.6 pg/mL of aflatoxin was reached within 53 min. *Pseudomonas aeruginosa*, a food-borne pathogen, was detected using SEF technique where (R)-4-(anthracen-9-yl)-6-(naphthalen-1-yl)-1,6-dihydropyrimidine-2-amine (ANDPA) as fluorescence probe and glucose stabilized silver nanoparticles (Glu-AgNPs) in various food samples including milk, orange juice and sugarcane (Ellairaja, Krithiga, Ponmariappan, & Vasantha, 2017).

Due to their specificity, immunosensors could be used in the agri-food sector for monitoring different types of molecules from pesticides (Miyake et al., 2019), bacterial and fungal toxins (Commission, 2006, p. 364; Z.; Mayer, Färber, & Geisen, 2003; Organization & Cancer, 1993; Park, Kim, Kim, & Ko, 2014; Patel, 2004; Squire, 1981), drug traces (Elliott et al., 1998; Lu et al., 2012; Smith, Ehrenfried, Dalidowicz, & Turberg, 2002), and allergens (Ashley et al., 2017; Pollet et al., 2011; Rebe Raz, Liu, Norde, & Bremer, 2010; Tomassetti et al., 2013; Yman, Eriksson, Johansson, & Hellens, 2006). SPR biosensors can detect a specific biomarker of a pathogen even in a fmol range (Bhunia, 2008; Rasooly & Herold, 2006; Vidic et al., 2013). Compared to conventional microbiological methods, plasmonic methodology was proved easiest to perform and more affordable (\$3 per sample was estimated for porcine circovirus 2 detection (Basso, Cruz, Silva, Pedrosa, & Araújo Junior, 2020)). Nevertheless, due to possible cross-reactivity of antibodies,

which could lead to false positive results, in some cases, DNA probes are preferred as recognition elements.

2.2. Plasmonic genosensors

Genosensors employ immobilized ssDNA or RNA probes as recognition elements and detect their hybridization with the target nucleic acid sequences (DNA-DNA or DNA-RNA). The design of robust DNA probes is needed to assure the maximal specificity for the target sequence and to prevent any cross-reactivity. The size of probes is a critical factor because long DNA sequences may be very efficient recognition elements in solution, but upon immobilization they may form secondary structures, and cannot access targets (Vidic et al., 2019; Vizzini, Braidot, Vidic, & Manzano, 2019).

Various DNA probes are available in literature for most prevalent food pathogens. For instance, DNA probes for *C. jejuni* detection were designed in the hippuricase gene (*hipO*) (Fontanot, Iacumin, Cecchini, Comi, & Manzano, 2014), *cadF* gene (Shams, Bakhshi, Moghadam, & Behmanesh, 2019), *PorA* gene (Gnanaprakasa, Oyarzabal, Olsen, Pedrosa, & Simonian, 2011) or 16 S rRNA gen (Vizzini et al., 2020). DNA probes for *Salmonella* spp. Selected from conserved Vi capsular antigen gene and the *invA* gene have been successfully applied in plasmonic and diffraction optics biosensors, respectively (Anu Singh, Verma, & Arora, 2015; D. Zhang et al., 2012). The sensor was built by the grafting of thiolated DNA detection probe on the gold surface via thiol-gold covalent bonds (Fig. 5A). Typically, such sensors can be re-used for dozens of hybridization cycles at room temperature without obvious loss of performance. They provide analysis within less than 1 h (Piliarik, Párová, &

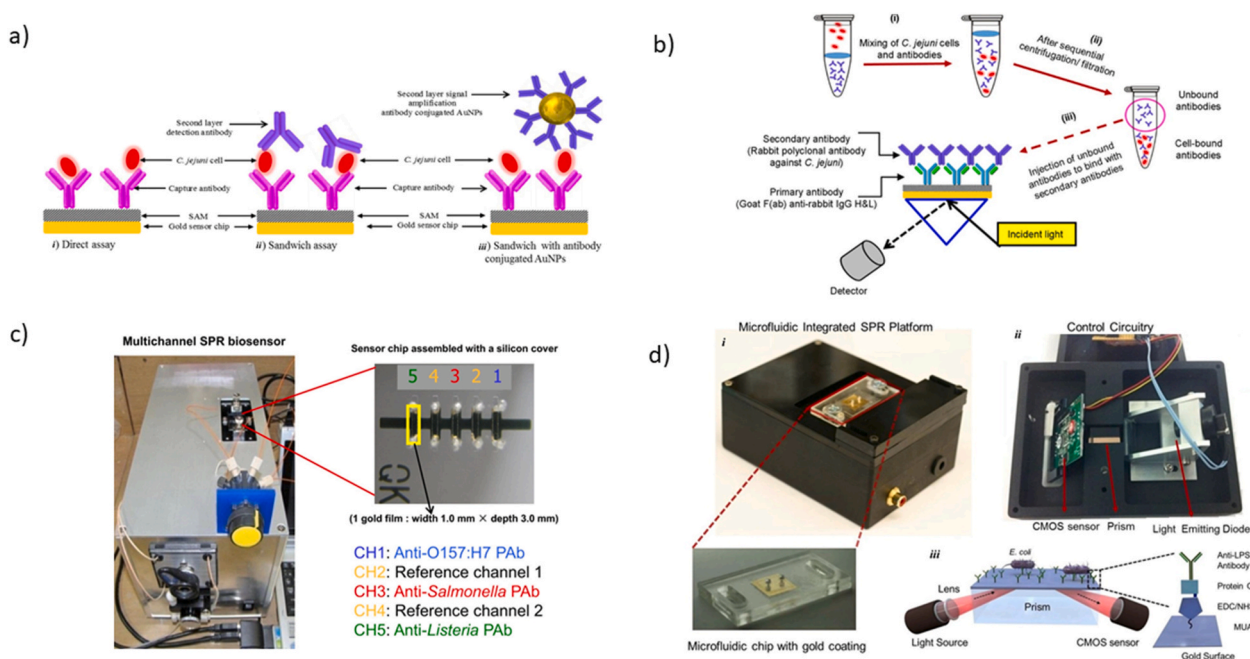


Fig. 4. Plasmonic immunosensors. (a) Detection of *C. jejuni* using three different formats: (i) direct; (ii) sandwich and (iii) sandwich assay coupled to antibody-functionalized Au NPs (adapted from (Masdor et al., 2017)). (b) Schematic presentation of the subtractive inhibition assay (adapted from (Masdor et al., 2019)). (c) Multichannel SPR biosensor based on sensing channels with immobilized antibodies that recognize *E. coli*, *Salmonella*, or *Listeria*, and two blank reference channels (adapted with permission from (X. Zhang et al., 2017)). (d) Portable plasmonic platform for pathogen detection and quantification with (i) integrated microfluidic chips and (ii) the SPR device that captures reflected light by a CMOS sensor. (iii) Schematics of the microfluidic integrated SPR platform. Adapted from (Tokel et al., 2015).

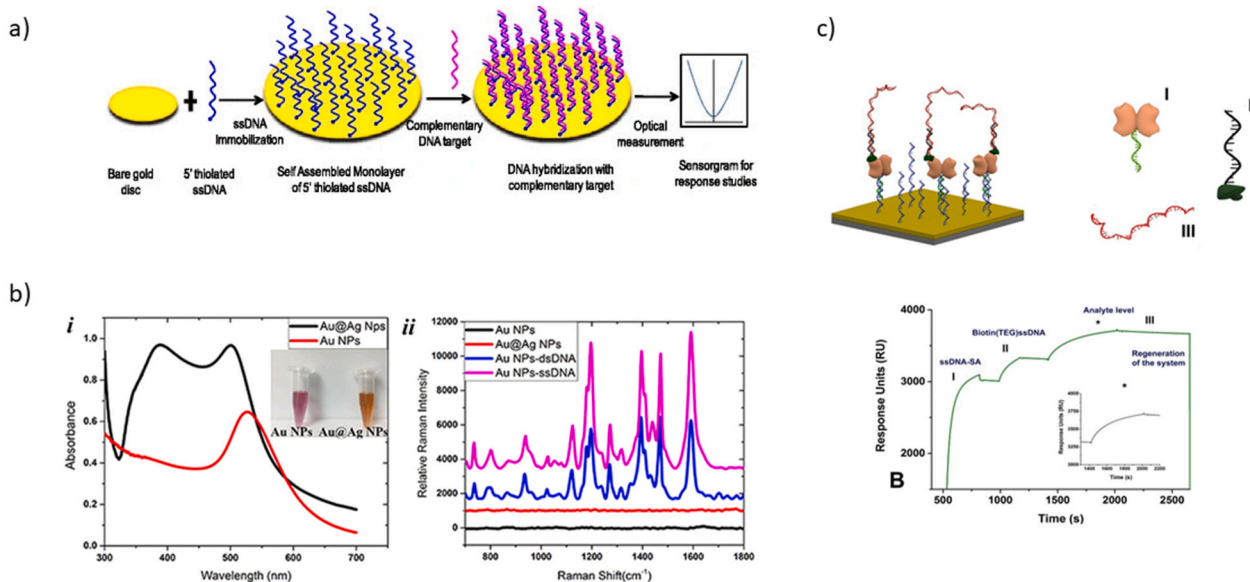


Fig. 5. Plasmonic genosensors. (a) SPR biosensor using DNA self-assembly (adapted with permission from (Anu Singh et al., 2015)). (b) UV/Vis absorption spectra and photos of Au NPs and Au@Ag NPs (i) and their SERS spectra before and after DNA binding (ii), adapted with permission from (Jiang et al., 2019)). (c) SPR biosensor construction using ssDNA-streptavidin (I), ssDNA-biotin detection probe (II) and target sequence (III). Low panel shows representative sensorgram of the capture concept. Adapted from (Plácido et al., 2020).

Homola, 2009) with the LOD similar to ones of PCR and qPCR analysis (pg/ μ L to ng/ μ L) (Manzano, Vizzini, Jia, Adam, & Ionescu, 2016; Shams et al., 2019). For instance, a DNA target of *Brettanomyces bruxellensis*, which produces unpleasant aromas in wine, was detected in wine matrices using LSPR with the LOD of 0.1 ng/ μ L (Manzano et al., 2016). Using an optimized SERS technique based on a nanostructured ultrafine glass support, *B. bruxellensis* genes were detected with the LOD of 0.1

ng/ μ L showing a high SERS sensitivity (Ionescu et al., 2020). A SERS based biosensors using double strand DNA bound bimetallic Au@Ag nanoparticles was developed for kanamycin detection in milk samples (Jiang et al., 2019). For this, kanamycin aptamer carrying the Raman reporter Cyanine-3 (Cy3) was hybridized with probe DNA embedded on the surface of Au NPs, and then nanoparticles were covered by Ag shells (Fig. 5B). Raman signals were stronger when kanamycin was bound to

the aptamer compared to the substrate itself. The sensor showed a broad linear range from 100 ng/mL to 10 µg/mL with an ultralow detection limit of 0.90 pg/mL. Furthermore, techniques like microspotting or microcontact printing provide a possibility to functionalize a sensor surface with various oligonucleotide probes for a multiplex pathogen detection without losing the sensitivity (Piliarik et al., 2009; Sikarwar et al., 2017; Vidic et al., 2007).

Detection of genetically modified organisms (GMOs) is another highly demanded application of plasmonic biosensors because of the strict legislations on GMO authorization in many countries. The genetic composition of a GMO has been altered through the insertion of a new gene or by deletion of an existing one, in order to increase productivity, resistance to pathogens or to environmental conditions (Cottenet, Blancpain, Sonnard, & Chuah, 2019). A renewable SPR biosensor chip for relative quantification of GMO soybeans enabled detection of the taxon-specific gene or the event-specific sequence with the LOD of 20 pM and 16 pM for roundup ready and lectin, respectively (Plácido, Ferreira-da-Silva, Leite, de-los-Santos-Álvarez, & Delerue-Matos, 2020). This label-free system enabled GMO quantification through the target DNA hybridization with the specific biotinylated ssDNA probes coupled to a SPR sensor chip via streptavidin-bound universal ssDNA (Fig. 5C).

The main weak point of genosensors is the demand of extraction and purification of genetic material prior to detection. The costs associated to sample preparation and bacterial enrichment are dominating in all actual food safety assays.

2.3. Other plasmonic sensors

Phage receptor binding proteins and aptamers are attractive recognition elements because they target whole bacterial cells which significantly simplify food analysis (Kotsiri et al., 2019; Vidic et al., 2017; Vizzini et al., 2019; Wolter & Mayer, 2017). Aptamers are single-stranded nucleic acid molecules that can selectively bind with a high affinity a broad range of targets. Aptamers can be easily chemically

derivatized to show extend lifetimes and bioavailability. With respect to antibodies, aptamers present lower costs, higher stability, easier modification and little or no batch-to-batch variation. Plasmonic aptasensors enable highly specific and sensitive detection of bacteria in food matrices as shown for *S. enteritidis* in chicken, pork and fish (Di, Du, Pan, & Wang, 2017), *L. monocytogenes* in food, environmental and clinical samples (S.-H. Lee et al., 2015), or *S. Typhimurium* in pork meat samples (Oh et al., 2017). For the sensitive detection of *L. monocytogenes* cells, an aptamer-based sandwich assay was constructed through immobilizing the capture aptamer onto the chip surface via amino-links, sample addition, and finally detection using a fluorescent-labeled detection aptamer (Fig. 6A). Detection can be coupled with gold nanoparticles for signal amplification as performed for the detection of *S. aureus* (Chang et al., 2013). A LSPR aptasensor was developed using nanosphere lithography to deposit a hexagonal array of Au nanotriangles onto a sensor chip (Hu, Fu, & Bohn, 2018). The surface was then modified with a sandwich of biotinylated polyethylene glycol, neutravidin, and biotinylated aptamers. This point-of-care device provided detection of *P. aeruginosa* in a dynamic range from 10^1 and 10^3 CFU/mL within less than 3 h.

In addition, aptamers selected to bind proteins and toxins can be applied for food analysis. For instance, a FO-SPR sensor was developed for the detection of Ara h1 and Ara h2 proteins that cause the allergy to peanuts (Tran et al., 2013). SPR aptasensors for detection of lysozyme, an additive used as a natural antimicrobial that causes the allergy to eggs (Mihai et al., 2015), tetracycline in honey (W.-Q. Li et al., 2016), ochratoxin A in wine and peanut oil (Z. Zhu et al., 2015), aflatoxin B₁ in moldy sugarcane (Tian et al., 2018), and avian influenza virus particle in duck feces (Kim et al., 2019) were reported. In another study, a highly sensitive detection of *E. coli* O157:H7 was achieved using SERS-aptamer based platform with the LOD of 10 CFU/mL in cell culture and 100 CFU/mL in ground beef samples (Díaz-Amaya, Lin, Deering, & Stanciu, 2019). The Raman probe-aptamers were covalently bound via 4-aminothiophenol to Au NPs. Specific capturing of bacteria led to cell

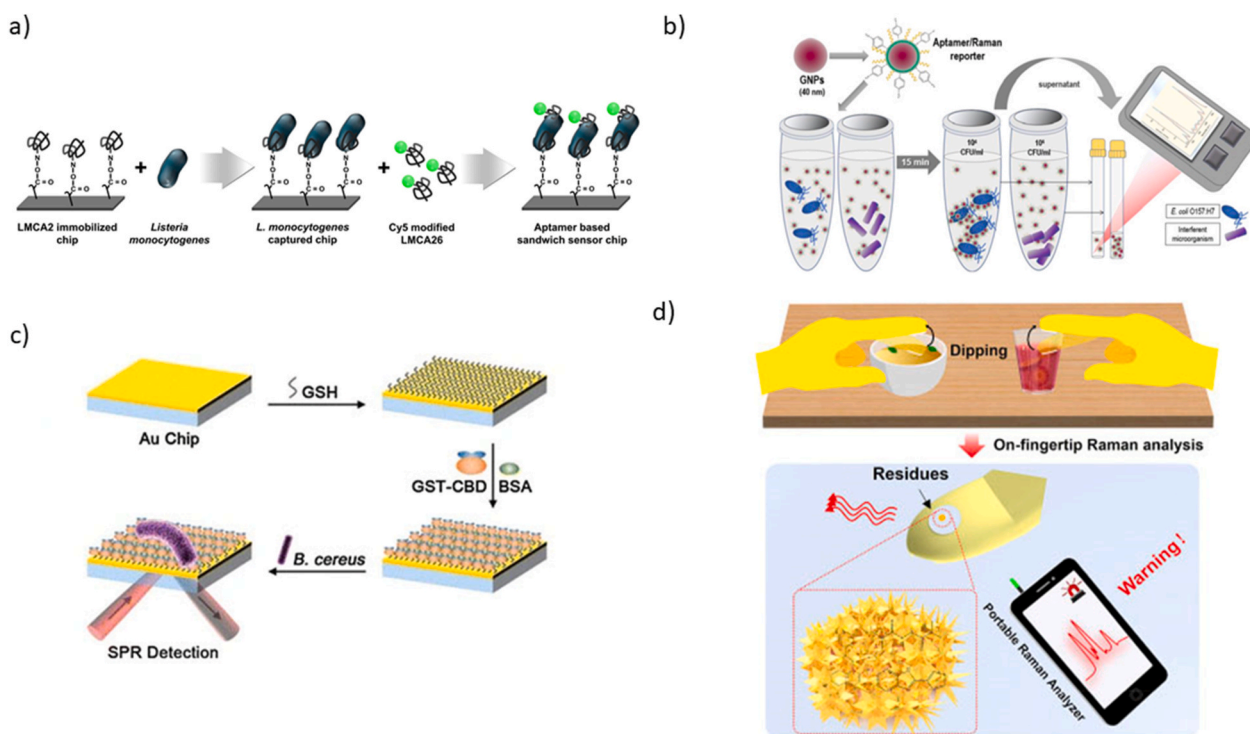


Fig. 6. Plasmonic apta- and phage-based sensors. (a) Aptamer-based sandwich assay, adapted with permission from (S.-H. Lee et al., 2015). (b) Aptamer-based whole cell detection of *E. coli* O157:H7. Adapted with permission from (Díaz-Amaya et al., 2019). (c) SPR detection based on a cell wall-binding domain of *B. cereus*-specific bacteriophage-modified chip. Adapted with permission from (Kong et al., 2015). (d) Design of tape-based SERS sensors for on-hand detection of food contaminants. Adapted with permission from (He et al., 2020).

sedimentation and the consequent formation of a superficial phase with unbounded Raman probes (Fig. 6B).

Phages and phage receptor binding proteins are alternative to aptamers and antibodies. Phages are bacterial viruses that possess consistency and withstanding harsh environmental resistance. Phages are unique bio-probes, owing to their selectivity, specificity to bind the bacterial host. SPR technique based on T4 bacteriophage that recognizes *E. coli* O157:H7 and BP14 bacteriophage that recognize methicillin-resistant *S. aureus* enabled these bacteria detection in less than 20 min, with a LOD of 10^3 CFU/mL without any labelling or enrichment procedure (Tawil, Sacher, Mandeville, & Meunier, 2012). Karoonuthaisiri et al. developed a SPR biosensor with the chimeric M13 bacteriophage as a probe that detected *Salmonella* spp. with the LOD of 3 CFU/25 g of food sample after a pre-enrichment step (Karoonuthaisiri et al., 2014). In another study, *Bacillus cereus*-specific bacteriophage endolysine PBC1 used in SPR biosensor detected *B. cereus* with the LOD of 10^5 – 10^8 CFU/mL in a direct format, and of only 10^2 CFU/mL in a subtractive inhibition assay (Kong et al., 2015). To immobilize the specific phage domain labeled with a GST tag (GST-PBC1) onto an Au chip, the surface was firstly coated with a self-assembled monolayer of glutathione (Fig. 6C) and albumin as a blocking agent. Recently, a high sensitivity was obtained by SERS technique using nanosculptured silver chips. The T4 bacteriophage was immobilized near to the plasmon surface allowing signal enhancement. A single cell of *E. coli* was detected and no cross reaction with control bacterial strains was observed (Srivastava et al., 2015).

A microdroplet-captured tape SERS platform based on highly branched Au nanodendrites that promotes SERS activity was developed for successful screening of food contaminants (He, Yang, Xu, Song, & Zhang, 2020). The tape-based sensors were attached on a wearable glove, and analytes were collected from solutions through a simple “dip and pull” procedure, leading to an on-site identification of residual contaminants (Fig. 6D). The sensor demonstrated an accessible signal response for food, forensic and environmental applications.

3. Discussion and future perspective

New strategies are needed to mitigate problems linked to the food contamination and to ensure the availability of an accessible, safe, nutritious, and abundant food supply. Biosensors could simplify procedure and radically reduce time, price and consumption of reactants, compared to traditional methods. Optical biosensors, in particular SPR, have been developed for detection of different targets, such as foodborne pathogens, pesticides, antibiotics, heavy metals, allergens, and toxins. In parallel, analytical improvements have been achieved by coupling different techniques (fiber optics, Raman, fluorescence, luminescence) to SPR in order to improve sensitivity. The future improvements will regard the miniaturization of instruments to handheld devices compatible with smart phone imaging (see Fig. 3), and simplification of analysis to enable direct target detection in food matrices. In particular, aptamer-based biosensors are interesting area to ensure food safety because they combine advantages of immunosensors (direct assay without DNA extraction and consequently reduction of analysis time) and genosensors (reduced cost and higher stability to temperature variations and chemicals). However, aptasensors show some limitations because they are sensitive to environmental conditions. Nanoparticles used to stabilize aptasensor may aggregate in biological solutions, which is additional challenge for improvements.

Despite the remarkable advantages of plasmonic biosensors, they still have some limitations. First, progress in surface chemistry and reliable immobilization of recognition elements are needed to decrease a non-specific binding and improve the efficiency of detection. Second, plasmonic sensors using nanoparticles have low sensitivity in comparison with surface plasmon sensors. Especially, LSPR sensor sensitivity is negatively affected by alterations in the shape of the LSPR peak when measuring the changes at the particle local surface environment. Third,

sample preparation and processing are still the limiting factors for achieving functional portable bio-sensors. Employing advanced microfluidic systems, which are already widely employed in other fields, is needed to enable automated sample handling in point-of-care and multiplex analytical applications in food analysis.

The main body of current research in plasmonic biosensors is devoted to generation of biochips that provide signal enhancement and increased stability. The plasmonic resonance shift can be increased by measuring a larger optical mass of target (which is obtained in a subtractive inhibition assay), or by incorporation of a sandwich mode that uses a chromophore with absorption in the visible region coupled with the plasmon resonance. Moreover, a high resonance shift and, thus, high sensitivity can be obtained by attaching different nanoparticles into the capturing layer. The nanoparticles for signal enhancement may be of different materials but monodisperse, because polydispersity will hinder spectral resolution. Beside the selectivity and specificity, the over-time stability, accuracy and reproducibility are still critical to achieve.

The possibility to perform tests within less than 1 h in a sensitive way will increase interest from the food industry and will positively affect the national food and agricultural systems by providing scientific evidences for decision-making. Over the last decade, the prevalence of foodborne diseases has been rising at high rate. In addition, there is an increasing need for the detection of new pathogenic agents in foods. Both of these factors are expected to drive the market for food security in the near future. Finally, while plasmonic technology is certain to have long lasting impact, the need for a simple and rapid food assay that does not require certified laboratories with complicated licensing requirements is pressing and guarantees the future development in this field.

Author contributions

Simone Balbinot and Anand Mohan Srivastav wrote the original draft. Jasmina Vidic, Abdulhalim Ibrahim and Marisa Manzano wrote, revised and edited this manuscript.

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Declaration of competing interest

Nothing declared.

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