Review article

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Coupling configurations between extended surface electromagnetic waves and localized surface plasmons for ultrahigh field enhancement

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Abstract: Local enhancement of electromagnetic (EM) fields near dielectric and metallic surfaces is usually associated with the existence of a confined EM wave at least in one direction. This phenomenon finds applications in enhancing optical spectroscopic signals, optical emission, nonlinear optical processes, biosensing, imaging contrast and superresolution, photovoltaics response, local heating, photocatalysis, and enhanced efficiency of optoelectronic devices. A well-known example is when the surface electromagnetic wave (SEW) is excited at the interface of two media, the field gets enhanced normally to that interface. This article reviews the different configurations revealing enhanced EM fields, particularly those giving ultrahigh enhancement, such as when a localized SEW is excited not from free space but via an extended SEW. Of particular interest are surface plasmon waves (SPWs) excited at the surface of metal-dielectric and particularly when exciting localized SPWs using extended ones. The latter case so far gave the highest local field enhancement; however, configurations involving Bloch SEWs, guided mode resonances, and cavity resonances have also been shown to give significant enhancement when used to excite localized surface plasmons. With this strategy, field enhancement by more than an order of magnitude can be attained. Using this ultrahigh enhancement, the strong coupling experiments between molecules and the intense optical field will be possible and new devices may emerge from those new methodologies for ultrahigh sensitive sensing for environmental and medical applications, as well as for improved optoelectronic devices.

Keywords: local field enhancement; ESP to LSP coupling; surface electromagnetic waves; plasmonic sensing; enhanced photocatalysis and energy harvesting.

1 Introduction and motivation

Nanophotonic and nanoplasmonic structures are becoming widely known in enhancing the efficiency of optoelectronic devices as well as in biosensing using enhanced absorption and scattering, refractive index changes, surface-enhanced Raman scattering (SERS), surface-enhanced fluorescence (SEF), and surface-enhanced infrared absorption (SEIRA) (see Figure 1) [1–23]. These applications of metallic nanostructures are attributed to their ability to generate intense electromagnetic (EM) fields at the nanoscale vicinity of their surface stemming from the surface plasmon resonances (SPRs). SPR is the coherent collective oscillation of free electrons at the metal surface with a momentum in resonance with the incident light momentum along the surface [24]. The motivation for further field enhancement schemes is clear due to its importance in many fields, both for research and practical products that may be built due to a large enhancement using low power laser and simple setups (Figure 1).

Generally speaking, there are two types of plasmons for noble metal nanostructures: propagating (or extended) surface plasmons (ESPs), also known as surface plasmon polaritons (SPPs), and localized surface plasmon resonances (LSPRs) [1, 24]. ESPs are usually supported by structures with at least one dimension approaching the excitation wavelength, for example thin metal films (<50 nm) attached to a prism (Kretschmann-Raether configuration) or on the surface of a bulk metal when a dielectric gap of the order of the wavelength or less is between the surface and the prism (Otto configuration). They travel along the metal-dielectric interface, and the fields of ESPs exponentially decay into both bounding media. In the case of LSPRs, plasmons are sustained by nanoparticles (NPs) much smaller than the incident wavelength. LSPRs

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Figure 1: Diagram illustrating the wide applications of local optical field enhancement.

Figure 2: General excitation schemes for extended and localized surface plasmons.

Scheme for exciting ESPs (A) through a coupling medium (prism, waveguide, fiber or grating) and for exciting LSPs (B) by direct excitation from free space. The wave is evanescent perpendicular to the interface, and therefore, the field is enhanced at the interface in both cases. The field penetrates the metal only within the skin depth, while in the dielectric, it can be from few hundreds of nanometers until a few microns in the ESP case and only to within a distance comparable to the radius of the NP in the LSP case.

of noble metal NPs give rise to intense absorption, scattering, and extremely enhanced EM fields at their resonance wavelengths. As seen in Figure 2A, the mostly used scheme for exciting ESPs is using incident transverse magnetic (TM) polarized light through a prism, waveguide, grating, or optical fiber (so-called coupling medium) on which a thin noble metal film (<50 nm) is deposited and the ESP is excited along the metal film-dielectric interface. The mostly used scheme for exciting LSPs (Figure 2B) is by directly

shining light on a collection of metal NPs on substrate or in solution. Contrary to the ESP case, in the LSP case, light does not need to be polarized as the excitation occurs due to the scattering, although for anisotropic particles, different polarizations can excite different LSP modes.

There are several methods for improving the enhancement factor of the EM field near a nanostructure, for example by tailoring the geometry, making sharp edges, and assembling different NPs in close proximity to each

Figure 3: Several factors affecting field enhancement upon plasmon excitation.

All parts A–E correspond to the titles on the left respectively. Designated figures were reproduced with permission from the following articles: (B) [6], (C) [25], (D) [26], (E) [27].

other (see Figure 3) [1, 2, 6–8, 10–18, 21, 28]. These are well-known techniques; however, here, our main interest is to highlight the less-known methodology, which is the excitation of LSPs not from free space but by ESPs generated as surface EM waves (SEWs) and excited from a special coupling medium (prism, waveguide, fiber, or grating). In the past decades, extensive research work has been done to explore metallic nanostructures that could generate hot spots with large EM field enhancement for high-performance sensors [29–35]. Particularly, the system composed of metal NPs positioned over a continuous metallic film has received increasing attention [25, 36–44]. In metal NP over the metal film (NPOMF) configuration, the NP couples with its mirror image in the metallic film, generating strongly enhanced EM fields localized at the junction between the NP and the metal film. The NPOMF can be fabricated using low-cost and simple methods over large areas by directly depositing the NPs on top of the metal film. In addition, the separation distance between the NP and the film can be well tuned by adjusting the thickness of the dielectric spacer, resulting in both the well-tuned resonance position of the supported NP and the highly uniform and reproducible hot spots. More importantly, NPOMF provides coupling in the vertical direction, for which uniformly distributed gaps between the NP and the film with sub-1 nm size can be fabricated. Such an extremely small gap is critical for

generating extremely enhanced EM fields in coupled nanostructures. All these advantages made the NPOMF a good platform for SERS, SEF, plasmon-enhanced photoluminescence, and other techniques.

Generally, the metal NP and the metal film can sustain LSPs and ESPs, respectively. For periodic NP array on metallic film, the NP array can excite the ESPs of the metal film by acting as a 2D grating to provide additional momentum resulting from the lattice. The generated ESPs at the metal-dielectric interface can then couple with the LSPRs or the Bragg waves of the NP array, generating higher EM field enhancement around the NPs [45–47]. However, the ESPs of the underlying metal film in NPOMF were seldom considered in these studies because ESPs cannot be directly excited using the free space incidence due to the momentum mismatch. Recently [48–50], some studies considered Au nanospheres over the Ag film as an example, in which a novel methodology was proposed to excite the LSP of the supported NPs using the ESP waves of the underlying metallic film. It was demonstrated that the excitation of LSPs using ESP waves can generate much higher EM field intensity than direct excitation of the LSPRs using incidence from free space, attributed to the strong confinement of the ESP waves in the vertical direction. First, the phenomenon was predicted theoretically using the finite-difference time-domain (FDTD) method, in which the largest

EM field enhancement is found to be generated when the incidence angle is the ESP resonance angle of the underlying metal film. In addition, the EM field enhancement was found to depend strongly on the dielectric spacer thickness when exciting the LSPs of the NP using ESP waves, showing a drastic decrease when gradually increasing the spacer thickness. Considering the matching between the LSPRs and the ESPs, the effect of the metal NP size on the EM field enhancement was also examined. Finally, the drastically enhanced EM fields generated by exciting the LSPRs using ESPs were demonstrated in SEF and SERS experiments [51]. The main aims in this review article are to highlight the main findings of recent works in this regard and propose future perspectives, particularly the possible extension of this ultrahigh enhancement scheme to other possible localized SEW excitations by other ESEWs and not necessarily limiting ourselves to the plasmons case.

2 The basic concepts of local field enhancement

The excitation of a surface wave causes confinement in at least one direction. Energy conservation then imposes concentration of the EM energy near that surface, thus the enhancement of the EM field. This is the essence of the plasmonic field enhancement near metallic surfaces. The enhancement association with the confinement maybe understood as a result of conservation of energy. Considering the energy Ξ associated with the electric field, it is proportional to the integral $\Xi = \int_{\Delta V} EE^* dV \approx \gamma |E|^2 \delta_x \delta_y \delta_z$ over the volume $\Delta V = \delta_x \delta_y \delta_z$ upon which the field extends, with γ being a proportionality constant and $\delta_x, \delta_y, \delta_z$ being the penetration depths of the evanescent field along each particular direction x, y , or z . According to this relation, as ΔV decreases when the wave is confined, the field intensity EE^* has to increase; it is a focusing mechanism and one can think of it as an uncertainty relation. Since enhancement in the ESP and LSP cases exhibit some differences, we shall treat each one separately in the following sections.

2.1 Improving the field enhancement in the ESP case

2.1.1 Prism coupling case

The simplest case is the ESP case described in Figure 2A, in which the SP wave is excited on the surface of thin

metal film through some coupling medium. Using the Shalabney-Abdulhalim algorithm [52], developed for calculating fields distribution in multilayered structures, it can be easily found that the field amplitude enhancement at the metal dielectric interface is given by the Fresnel transmission coefficient $|t|$ through the metal layer for the magnetic field case H_y and by $|t \cos \theta_m / n_m|$ for the electric field E_x . Several approximations were done in the scientific literature [53, 54] to extract a simplified analytic expression for the field enhancement for the ESP case and show that there is a proportionality to the ratio between the real and the imaginary parts of the metal dielectric constant: $|\epsilon_{mr}^\alpha / \epsilon_{mi}^\beta|$, where approximately $\alpha \approx \beta \approx 1$. Hence, the higher this ratio is, the higher is the enhancement factor. Physically, this can be understood as a result of lower dissipation in the metal surface. Several prism coupling configurations were investigated by Shalabney and Abdulhalim [52], showing a direct correlation between the sensitivity of the SPR to the analyte refractive index and the field enhancement. As the SPR angle increases, for example when using a lower index prism or adding a nanolayer of high index material on top of the metal, the field enhancement also increases. Figure 4A shows the enhancement at different incidence angles inside the prism showing the drastic enhancement at the resonance angle. The shape and width of the enhancement factor versus angle are the same as the SPR reflectivity shape and width.

Figure 5A presents the field distribution for different prism refractive indices. As mentioned, increasing the prism refractive index decreases the incidence angle required inside the prism for the SPR excitation condition. Physically, it can be understood as a result of the increase in the field component normal to the interface E_z as the incidence angle increases. The normal field component is responsible for generating the surface charge. Figure 5B shows the field distribution at different wavelengths, showing that it decreases with the wavelength partially because the ratio $|\epsilon_{mr}^\alpha / \epsilon_{mi}^\beta|$ decreases.

Another methodology for varying the material properties is by controlling the porosity of thin metal films. This can be done using the oblique angle deposition technique, in which the shadowing effect causes a columnar structure to be formed. The effect of the porosity of such metal films on the SPR signal, field enhancement, SERS, and SEF was studied by several groups [6]. As the porosity increases, the metal is expected to be less absorptive; that is, the imaginary part of the dielectric function becomes smaller, and therefore, the enhancement near the metal film in the Kretschmann configuration is expected to get enhanced. This is demonstrated in Figure 6 showing the

Figure 4: Simulated field component (x and z) intensity distribution for the case of silver film on top of a prism and water analyte adjacent to the silver as a function of the distance from the prism interface: (A) $|E_x|^2$, (B) $|E_z|^2$. The parameters are shown on the top and the resonance angle obtained with the structure is 54.61° . Reproduced with permission from Reference [55].

Figure 5: Simulated x -component of the electric field distribution for silver film on prism configuration with water as analyte, $n_a = 1.33$. (A) At different prism refractive indices with metal layer thicknesses d_m that were chosen to obtain resonance for each prism, refractive indices are 41.5, 47.5, and 47 nm for 1.41, 1.61, and 2.5 prism refractive indices, respectively. (B) For different wavelengths at the resonance with the same metal thicknesses d_m for obtaining resonance in each wavelength, refractive indices are 47, 43.25, and 29.75 nm for 632, 850, and 1550 nm, respectively, and fixed prism index $n_p = 1.732$. Reproduced with permission from Reference [52].

larger enhancement as the porosity increases. Above 35% porosity, the angular SPR curve becomes very wide, indicating that more localized surface plasmons (LSPs) start to take effect, which is consistent with the fact that above a certain percolation threshold, the film becomes non-conductive. At the percolation threshold, there is a jump usually of the dielectric function, which might cause a jump in the field enhancement near the nano-columns. Following SERS and SEF investigations, there is indeed an optimum of the signal enhancement around 35% porosity.

Other methods for enhancing the local field further in the ESP configuration include the addition of a nanolayer on top of the metal film. With the top nanolayer having thickness below the cutoff for exciting guided modes and high refractive index, one can get enhancement by nearly an order of magnitude (Figure 8). As it is shown in Figure 7A, the field intensity enhancement is by almost an order of magnitude ($\times 9$) between the optimum case of having 10 nm top layer of Si and the case of no Si layer. On the other hand, in Figure 7B, the enhancement is

Figure 6: Simulated field distribution for thin metal on prism configuration with different porosities of the columnar silver film. Wavelength used is 1550 nm. Reproduced with permission from [6].

smaller ($\times 5$) and the reason is because in case (a), a prism of high refractive index is used, so to begin with, the field enhancement was already smaller by a factor of 2 than the case in (b). Note that the optimum layer thickness is 10.5 nm for Si (it depends on the refractive index of the material used) and it is much lower than the cutoff for guided mode excitation (~ 70 nm for Si). This is why this configuration is called nearly guided wave resonance, to distinguish from the case of a thick enough top dielectric layer that allows the excitation of guided modes. The addition of the nanodielectric layer shifts the SPR to higher angles, and therefore, in a similar manner to the effect of the prism refractive index (Figure 5A), one expects the field enhancement to increase [56]. Physically, it can be understood as a result of the increase in the field component

normal to the interface E_z as the incidence angle increases. The normal field component is responsible for generating the surface charge. Another observed phenomenon is the shift of the field maximum point from the metal surface to the Si interface with the analyte. This shift may be understood as a result of the formation of surface charge on the second Si layer interface when the plasmon is excited. The higher the dielectric constant is, the higher is the surface charge, which explains why Si gives higher enhancement than lower-refractive-index materials do.

2.1.2 Grating coupling case

In grating coupling, the additional momentum required to excite the ESP is provided by the wave vector of the grating: $2\pi m/\Lambda$, with m being an integer and Λ being the grating period. Several grating configurations have been studied, while here we limit ourselves to those on which field distribution calculations were highlighted: (i) surface relief metal grating on bulk metal [57], (ii) combination of thin metal grating with thick dielectric grating [58], (iii) combination of thin dielectric grating with thin metal film [59, 60], (iv) periodically perforated nanoscale spaces (holes or slits) yielding what is called enhanced optical transmission (EOT) [61–68], and (v) combination of multiple dielectric or metal gratings and metal film [69–78]. There are many works on grating-based SPR sensors [79, 80]; however, the field distribution calculations were absent from many of these works, and in some works, they are presented in a normalized fashion, so it is difficult to estimate the enhancement factor. Usually, with grating-based structure, those presented show very high field enhancement, particularly near the corners of the metal gratings with rectangular shape. Figure 8 shows the field enhancement

Figure 7: Field intensity distribution for the case of having a nanolayer with high refractive index on top of the metal film. (A) Geometry of the structure and field distribution for the case of $\lambda = 653$ nm, $n_p = 1.77641$, $n_{Si_film} = 3.881 + 0.0116i$, $n_{water} = 1.33$, $d_{Ag} = 43$ nm, $d_{Si_film} = 10$ nm. (B) Field intensity distribution at different Si layer thicknesses for the case $\lambda = 633$ nm, $d_m = 43$ nm, $n_p = 1.732$, $\theta_r = 54.61^\circ$, $n_a = 1.33$. Reproduced with permission (A) from [55] and (B) from [52].

Figure 8: Illustration of the field enhancement in grating configurations upon exciting SPR and GMR.

(A) Scheme of surface relief silver grating at oblique incidence excitation and the electric field intensity distribution at the ESP resonance for (B) when the first-order diffraction beam $m=-1$ excites the ESP and (C) when the second-order diffraction beam $m=-2$ excites the ESP. (D) Geometry of the metallic grating coupled with GMR structure, and the magnetic field amplitude distribution along the y direction at the ESP resonance (E). Parts A–C are reproduced with permission from [57], while parts D–E are from [58].

for two cases, the first a surface relief grating (Figure 8A) and the second a metallic grating adjacent to guided mode resonant structure (GMR) (Figure 8B). In both cases, the enhancement is larger or comparable to the prism coupling case by 1–2 orders of magnitude, and at the corners of the grating lines, it can even approach 3 orders of magnitude. At the corners, the large enhancement is due to the excitation of LSPs. In fact, it can be considered as another case of the main topic of this article – ultrahigh enhancement configuration, in which the coupling is between the ESP generated due to the periodicity in the lateral dimension, which in turn excites the LSP on the corners. We will come back to this point later in the manuscript.

A newly investigated grating coupling configuration is the one shown in Figure 9A, in which a thin dielectric grating is used on top of a thin metal film [59, 60]. This configuration was shown to allow the excitation of two ESPs, on either side of the metal films (analyte mode and substrate mode). As the analyte refractive index changes, it shifts to a different wavelength or angle while the substrate mode is nearly fixed so it can be used as a reference and vice versa (Figure 9B). The field intensity distribution shown in Figure 9C and D exhibits high field enhancement near the interface. Note that for the substrate (reference) mode, the field penetration depth is larger, indicating a

long-range SPR (LRSPR) analogous to the case of LRSPR excited in the prism configuration when an under-layer dielectric film is deposited between the prism and the metal film.

The idea behind the configuration presented in Figure 9 was stimulated from the case of thin metal gratings with spaces between the lines of the order of tens of nanometers. This structure is well known to reveal EOT at specific wavelengths that correspond to the dual ESP excitation by the grating [66, 81–84]. Similarly, two-dimensional (2D) gratings or arrays of nanoholes are known to exhibit a similar behavior. When the metal is thin enough, such as 20–40 nm of silver or gold, the transmission increases, but an additional ESP transmission peak appears that corresponds to an ESP generated at the interface with the substrate. Similar to the previous case, the substrate mode can be used as a reference for monitoring variations in the analyte concentration. Figure 10A shows the grating geometry, spectral transmission (B), and the field intensity distribution for each mode (C–D). Again, the field enhancement increases drastically at the resonance, with maximum values near the line corners. Note also that the substrate mode has a larger penetration depth indicating long range surface plasmon excitation.

Figure 9: Field enhancement upon double plasmon excitation using thin dielectric grating and thin metal film.

(A) Schematic of thin dielectric grating on top of a thin metal film configuration. (B) Reflectivity spectrum at normal incidence exhibiting analyte and substrates modes (SPR and LRSPR). (C) and (D) Electric field intensity distribution at the wavelengths of the two excited ESP modes. It should be noted that the field distribution for this geometry presented in [59, 60] is correct but the scale had an error in these references.

2.2 Improving field enhancement in the LSP case

For the LSP excitation on the surface of a single metallic NP, the anisotropic Mie theory predicts the following enhancement factor for the field amplitude: $|\varepsilon_{mr}/L_j\varepsilon_{mi}|$, which again shows that the ratio between the real and the imaginary parts of the dielectric constant plays a key factor. However, here, there is an additional important factor, called the geometrical factor L_j , which depends on the shape of the NP, and assuming a rotation ellipsoid, the index j designates the direction of one of the principal axes of the ellipsoid. To see this, we may write the field near the surface of a small NP as the sum of the incident field and the field from the polarization induced in the NP, a type of Mossotti equation in a tensorial form:

$$\vec{E} = \vec{E}_{inc} + 4\pi(1 - \tilde{L})\tilde{\alpha}\vec{E}_{inc}. \quad (1)$$

Here, L is the geometrical factor tensor and α is the polarizability tensor per unit volume. This equation agrees well with that derived by Tanabe for the maximum field in nanoshell-type particles [85]. The field intensity enhancement factor along any one of the axis j of an ellipsoid may then be written as

$$\eta_{E_j} = \left| \frac{E_j}{E_{inc}} \right|^2 = \left| 1 + (1 - L_j) \left(\frac{\varepsilon_m - \varepsilon_a}{L_j(\varepsilon_m - \varepsilon_a) + \varepsilon_a} \right) \right|^2 = \left| \frac{\varepsilon_m}{L_j(\varepsilon_m - \varepsilon_a) + \varepsilon_a} \right|^2. \quad (2)$$

Here, ε_m , ε_a are the dielectric constant of the metal and the surrounding analyte.

$$\eta_{E_j} = \frac{\varepsilon_{mr}^2 + \varepsilon_{mi}^2}{|L_j(\varepsilon_{mr} - \varepsilon_a) + \varepsilon_a|^2 + |L_j\varepsilon_{mi}|^2} \quad (3)$$

At the resonance, $|L_j(\varepsilon_{mr} - \varepsilon_a) + \varepsilon_a| = 0$, leading to

$$\eta_{E_j}^{\max} = \frac{\varepsilon_{mr}^2 + \varepsilon_{mi}^2}{L_j^2\varepsilon_{mi}^2} \approx \left(\frac{\varepsilon_{mr}}{L_j\varepsilon_{mi}} \right)^2. \quad (4)$$

For a sphere, $L_j = 1/3$, and we found that for a Ag sphere, the maximum intensity enhancement factor at 370 nm is around 70, and for a Au sphere, it is around 25 at 570 nm wavelength, in agreement with rigorous calculations.

The high enhancement near sharp edges or nanotips may be explained based on the geometrical factor effect. Hence, two main effects play a key role in determining

Figure 10: Field enhancement in the case of double SPR excitation using thin periodic array of metal nanoslits under EOT. EOT grating geometry (A), spectral transmission (B), and the field intensity distribution for each mode (C and D). Grating parameters are shown in the figure. Figure 10B is reproduced with permission from Reference [84].

the field enhancement factor: the material properties and the geometrical shape. Through the years, researchers have looked for additional ways to enhance the local field further, and one of the methods is based on interference effects, which can be implemented in several ways. For example, having two or more particles in close proximity to each other, the EM fields between the NPs interfere and generate hot spots with much larger local intensities than that of the individual fields. Interference effects take place also in submicron and micro-rods, and when the rod length is an odd multiple of the effective wavelength inside the rod, it acts as a nanoantenna, producing resonances with large enhancement in the infrared range [86–91] (see Figure 3). In Figure 11, a selection of field intensity distributions around NPs and NP arrangements that show high enhancement is presented demonstrating the main effects responsible

for the enhancement mentioned above. As it is seen, the field intensity enhancement is in the order of 100–600 for the majority of cases and sometimes it can arrive to the range of 1000, particularly at very localized hot spots. The coaxial nanoantenna enhancement is particularly remarkable in the high enhancement obtained over a wide spectral range, important for solar energy harvesting.

2.3 The basic concepts of the coupling and ultrahigh field enhancement

The basic concept is based on exciting LSPs from ESEWs and more particularly ESPs, which may be understood based on Figure 12. On the left side, the LSPs are excited from free space, while on the right side, they are excited

Figure 11: Selection of field intensity distribution calculations for different types of NPs and NP arrangements.

(A) and (B) triangles excited at different orthogonal polarizations, (C) nanorod, (D) nanoellipsoid; (E) bowtie; (F–G) disc in ring microresonator; (H–I) coaxial nanoantenna. Figures are reproduced with permission from (A–D) [92], (E) [93], (F–G) [94], 2007, and (H–I) [27].

via an ESP wave. Under the exact matching of the excitation conditions, one can roughly estimate the field enhancement factor to be determined by the product of the two separate enhancement factors: $F_c = F_{esp} F_{lsp}$, the ones associated with the ESP (F_{esp}) and the LSP (F_{lsp}), respectively.

In reality, an even higher enhancement than this was obtained, which is believed to be due to the fact that when the NP is close to the metal surface, its response changes due to the mirror charge. As it will be shown later when the extended surface electromagnetic wave (ESEW) is not

an ESP wave, the enhanced factor is less than the product above. One can think then of many other configurations such as waveguide coupling, grating coupling, fiber coupling, and a variety of NPs (see Figure 13). ESEWs can also be generated [95] using periodic structures (Bloch and Tamm waves) or absorbing layers (Zenneck waves), as will be shown later.

In a good approximation, the coupling between the two surface waves may be treated similar to the system of two coupled oscillators with two Eigen-frequencies $\omega_{1,2}$ coupled together with a coupling constant κ (see

Figure 12: Schematic to explain the main concept for the ultrahigh enhancement geometry excitation of LSPs via (A) plane waves and (B) extended SPWs.

Figure 14) [96–99]. In classical oscillators with masses and spring constants $m_{1,2}$, $k_{1,2}$, the Eigen-frequencies are given by $\omega_{1,2} = \sqrt{k_{1,2}/m_{1,2}}$. The equations of motion may then be written as

$$\ddot{q}_1(t) + \gamma_1 \dot{q}_1(t) + \omega_1^2 q_1(t) + \kappa \dot{q}_2(t) = \eta E(t) \quad (5)$$

and

$$\ddot{q}_+(t) + \gamma \dot{q}_+(t) + \omega_0^2 q_+(t) + \kappa \dot{q}_-(t) = \eta E(t) \quad (7)$$

$$\ddot{q}_-(t) + \gamma \dot{q}_-(t) + \omega_0^2 q_-(t) - \kappa \dot{q}_+(t) = \eta E(t). \quad (8)$$

Figure 13: Several configurations for exciting LSPs via ESPs. (A) prism coupling, (B) fiber coupling, (C) waveguide coupling, and (D) grating coupling.

$$\ddot{q}_2(t) + \gamma_2 \dot{q}_2(t) + \omega_2^2 q_2(t) - \kappa \dot{q}_1(t) = 0. \quad (6)$$

Here, $\gamma_{1,2}$ are the damping constants and the driving force for the first oscillator is the one generated by the incident time harmonic optical field $f(t) = \eta E(t) = \eta E_0 e^{-i\omega t}$ with some efficiency factor η .

The coupled two-oscillator Equations (6) and (7) were used to explain many phenomena in physics, including EM-induced transparency, plasmon-induced transparency, Fano resonances both in quantum mechanical systems and classical systems, as well as coupling between plasmons and molecules [96–99].

One of the intriguing results is that the system of two oscillators has two normal modes expressed in the splitting of the plasmon dispersion relation, which is maximized under the condition of exact coupling or zero detuning, $\omega_1 = \omega_2$. The two mode solutions can be seen easily if we subtract and add the two Equations (5) and (6) under the exact matching condition and same damping constant:

Figure 14: Schematic of two coupled oscillators each at its Eigen-frequency while only one of them is driven by an external force.

Here, $q_{\pm} = q_1 \pm q_2$, meaning that one mode is when the two masses oscillate in phase and the second is when in anti-phase. In the absence of damping, the two normal modes have the following frequencies:

$$\omega_{\pm} = \frac{\omega_{LSP}}{2} + \frac{\omega_{ESP}}{2} \pm \frac{1}{2} \sqrt{A + (\omega_{LSP} - \omega_{ESP})^2}. \quad (9)$$

Here, $A = \sqrt{\frac{N}{V}} \frac{e}{\sqrt{\epsilon_0 m}}$, where N/V , e , m are the volume concentration of oscillators (NPs in our case), electron charge,

and mass, respectively. The splitting at the resonance is given by the Rabi frequency:

$$\Omega = \omega_{+} - \omega_{-} = \sqrt{A}. \quad (10)$$

Using Drude-type damping, the normal modes become

$$\omega_{\pm} = \frac{\omega_{LSP}}{2} + \frac{\omega_{ESP}}{2} - \frac{i\gamma}{4} \pm \frac{1}{2} \sqrt{A + \left(\omega_{LSP} - \omega_{ESP} + \frac{i\gamma}{2} \right)^2}. \quad (11)$$

And the Rabi splitting

$$\Omega = \omega_{+} - \omega_{-} = \sqrt{A - \frac{\gamma^2}{4}}. \quad (12)$$

Hence, the Rabi splitting decreases as the damping increases. The strong coupling (Rabi splitting) in the dispersion curve is indeed observable only when the damping rate of the individual coupling states is smaller than the

Figure 15: ESP-LSP coupling in prism configuration and Rabi-splitting.

(A) Schematic representation of the experimental setup used to measure reflections in the Kretschmann configuration. Reflection measurements are made over 40° – 50° with $\sim 0.2^{\circ}$ angle increments. (B) Schematic representation of the ESP-LSP coupling. (C) SEM images of uniformly coated Ag NPs on 3-aminopropyltriethoxysilane (APTES) modified silicon substrate. The inset indicates an AFM image of the Ag NPs. (D) Extinction spectrum of Ag NPs. (E) Reflection spectra obtained from a bare Ag thin film and Ag NP-coated Ag film at around 45° incidence angle. The upper and lower polariton bands are at ~ 525 and ~ 650 nm, respectively. Figure and caption reproduced with permission from [100].

Rabi splitting. In spectroscopy, this means that the spectral width of each spectral line of the two oscillators is less than the Rabi splitting. Experimental verification of plasmon splitting due to ESP-LSP coupling and detailed investigation were performed in [100–102] (see Figure 15). This treatment gives an insight into the related physics; it does not give the field enhancement factor as a result of this coupling. For this rigorous EM, simulations are needed.

3 Examples of ultrahigh field enhancement

3.1 ESP-LSP coupling in Kretschmann-Raether configuration

The simplest case is the use of the prism configuration with metal NPs dispersed on top of the metal film (Figure 16A). The calculated EM field intensity distributions are shown in Figure 16B at different incidence angles, showing that it is few orders of magnitude higher at the ESP resonance angle 53.775°; it seems that the NP is “ignited” at the resonance angle. As shown in Figure 1B, EM fields are dominantly confined at the gap region between the Au nanosphere and the Ag film when the incidence angle

is away from the ESP resonance angle. The strongly confined and enhanced EM fields at the junction between the metal NP and metallic film at off-resonance angles have been explained by the near-field coupling between the NP and its mirror image in the metallic film [25, 36–40]. When gradually adjusting the incidence angle to approach the ESP resonance angle (53.775°) of the Ag film, the EM fields within the gap region become more and more intensely enhanced. At the same time, hot spots with large EM field enhancement are distributed not only at the gap region between the Au sphere and the Ag film. Note that the field distribution is asymmetric because the light is incident from the left side. The full width at half maximum of the maximum field versus incidence angle curve (Figure 16C) is comparable to the same angular width of the SPP wave on the surface of the silver film.

When the incidence angle is decreased to 45°, the maximum EM field enhancement is only 994, becoming smaller by 250 times than the value at the resonance angle. The EM field enhancement of 2.5×10^5 obtained through exciting LSPs using ESP waves shown here is much larger than that achieved in previous studies, where the maximum EM field enhancement for metal NPs over a glass substrate with a 2-nm separation is typically $\sim 10^2$, and the maximum EM field enhancement for metal NPs over a metal film with a 2-nm gap at off-resonance conditions is

Figure 16: Simulated field enhancement in the ESP-LSP coupling in prism configuration versus incidence angle.

(A) Schematic illustration of an Au nanosphere over an Ag film separated by a thin Si spacer. The silver film is coated on a SF-11 prism in the KR configuration. θ is the incidence angle in the prism at wavelength 655 nm. (B) Electric field distributions. (C) Maximum electric field enhancement. An Au sphere of 40 nm diameter is used on 46.4 nm Ag film with 2 nm Si spacer with different incident angles θ . The Ag film is coated on a SF-11 prism. The surrounding medium is water. The scale bar in each figure of (B) is 20 nm. In this study, all the calculations are performed using the FDTD simulation program (FDTD solutions 8.6, Lumerical solutions, Inc., Vancouver, Canada). Reproduced with permission from [48].

Figure 17: Spacer layer effect under ESP-LSP coupling in prism configuration.

(A) Schematic illustration of a 46.4 nm Ag film deposited on a SF-11 prism in KR configuration. The Ag film is covered with 2 nm Si layer. (B–D) E_z distributions of the structure shown in (A) with 45° , 53.775° , and 60° incidence angles. The color bar applies to all the EM field maps. (E) Electric field distribution of a 40 nm Au nanosphere directly deposited on a SF-11 prism without Si spacer and Ag film. The wavelength is 655 nm and the incident angle is 53.775° . (F–G) Calculated electric field distributions of a 40 nm Au sphere on top of a 46 nm Ag film at 532 nm wavelength with 37.25° and 60° incident angles. The Ag film is deposited on a SF-11 prism.

typically $\sim 10^3$ or $\sim 10^4$ [25, 36–40]. The field distribution of each part of the nanosystem gives hot spots of the order of 100 for both ESP with 2 nm Si on top of the silver film and an Au sphere of 40 nm diameter on top of the SF11 prism (Figure 17A–E). The multiplication by the two factors gives total enhancement of around $F_c = F_{\text{esp}} F_{\text{lsp}} \approx 10^4$, which is less than the one obtained (2×10^5) when the coupling occurs by at least an order of magnitude. This may be understood as a result of the interaction between the NP and metal film via the mirror charge. In this case, excitation of the LSP from free space reveals field enhancement factor of the order of $10^3 \sim 10^4$ at the resonance as seen in Figure 17F–G, thus yielding $F_c = F_{\text{esp}} F_{\text{lsp}} \approx 10^5 \sim 10^6$.

Optimization of the ESP-LSP nanosystem for field enhancement is highly important for surface-enhanced spectroscopy experiments (Figure 18A). First, the NP diameter is optimum depending on the wavelength; for example, for AuNP at 532 nm, excitation the optimum diameter is around 50 nm, while for 785 nm excitation, it is around 100 nm diameter. One of the important parameters is the vertical gap between the NPs and the metal surface. This was shown to be optimum when the NP is as close as possible to the metal surface. However, a certain gap is usually introduced for two reasons: (i) when the NP is closer than 2 nm distance from the metal, surface charge tunneling might occur, and the simulation does not take it into account, and (ii) a single molecular layer exists usually between the NPs and the metal surface for specific binding in biosensing applications or to demonstrate surface enhancement of spectroscopic signals. Therefore, a 2-nm gap is usually introduced with a refractive index of 1.6. The reason for using a Si gap in the simulations of Figure 15 was to check possible further enhancement of the field because it is known that the ESP field gets enhanced when 10 nm of Si is added to the silver film [103,

104]. It was found that this is not the case, and the reason is possibly that the LSP field becomes very small as a gap is introduced between the NP and the metal layer. Another important optimization parameter is the lateral or horizontal gap between the NPs. At first sight, one may expect to get higher enhancement as the NPs become closer because it is well known [105] that as two NPs get closer to each other, hot spots between them may arise with large field strength. The rigorous simulations showed an opposite behavior and the field enhancement increases, with the horizontal gap reaching a saturation level above a distance around 1500 nm (Figure 18B).

This may be explained based on the two oscillator models mentioned previously. Within this model, the splitting is proportional to the number density of the NPs, $\sqrt{N/V}$. Hence, getting the NPs together causes the splitting between the two Eigen-frequencies to increase, meaning now, the excitation frequency is more far from each one of the Eigen-frequencies, and therefore, the coupling becomes less efficient. Therefore, the field enhancement decreases as the NPs become closer, contrary to the Rabi-splitting behavior. Above a certain distance (~ 1500 nm in this case), the interaction between the NPs becomes weak and they start to behave as single isolated particles. One may state then that for the ultrahigh enhancement of the local EM fields, it is better to be in what is called the “weak” coupling regime. This behavior is shown in Figure 18B, left axis; however, for surface-enhanced spectroscopy, the incident beam is usually large and covers many hot spots. As a result, one should consider the average field, which also shows maximum around the same horizontal distance but then starts to decrease. To confirm these findings experimentally, the SEF (Figure 18C) and SERS (Figure 19) signals were measured as described in the configuration of Figure 18A. The inter-particle distance was changed

Figure 18: SEF under ultrahigh enhancement with prism configuration.

(A) Schematic of the experimental setup used in the ESP-LSP coupling experiments and the measurement of the surface enhanced spectroscopy signals. (B) Effects of the horizontal gap “ g ” between two Au NPs on the maximum (left) and average (right) electric field enhancement in Au NP arrays over the Ag film at 785 nm with 34.645° incidence angle inside the prism. (C) Fluorescence intensity of a monolayer of Rhodamine 6G covered on a 47 nm Ag film with various concentrations of 40 nm Au nanoparticles deposited on the Ag film at 532 nm. The low intensity signals between 640 nm and 680 nm are zoomed in and shown in the inset. The incidence angles for the on- and off-resonance conditions are 37.25° and 60° , respectively. Reproduced with permission from (A–B) [51] and (C) [48].

Figure 19: SERS under ultrahigh enhancement with prism configuration.

SERS spectra (A) for on ESP resonance at Ag/4ATP configuration and on and off ESP resonances at the optimum Ag/4ATP/AuNP structure and (B) from the Ag/4-ATP/80 nm AuNP structure with varying AuNP solution concentrations. The inset shows the variation of SERS peak signal at 1077 cm^{-1} with AuNP concentration. Excitation wavelength is 785 nm. Reproduced with permission from [51].

Figure 20: FDTD simulation results of the field intensity distribution around a gold nanorod of 50 nm diameter and 300 nm long on top of silver film on SF11 prism for the ESP-LSP coupling in the near infrared (1500 nm wavelength) at different orientations of the nanorod. The angle inside the prism is 50.764° and dielectric spacer layer is 2 nm of $n=1.6$ between the NP nanorods and the 32-nm-thick Ag film. The light is incident from the left side.

by spinning NPs with different concentration, and the optimum concentrations were found to reveal average distance not far from the theoretically expected one.

Particle shape, arrangement on the metal surface, and materials are other parameters that one can optimize to get larger enhancement in the particular wavelength range of interest. As an example, the use of nanoantenna on top of the metal surface can boost the field enhancement in the infrared range. In Figure 20, one example of Au nanorods (AuNRs) in the form of cylinders having 50 nm diameter and 300 nm length are deposited on top of a silver film at different inclination angles. The light is incident from the left side, which explains the asymmetry in the field distribution. The AuNRs are designed to act as nanoantenna in the short wave infrared (SWIR) range. It is interesting that the maximum field has some dependence on the inclination angle peaking at 60° but it falls within the range 10^5 – 10^6 , which is much higher than regular field enhancement factors obtained by direct excitation. This fact relaxes the tolerance on the deposition conditions of AuNRs. One of the possible ways to deposit such AuNRs is using the glancing angle deposition (GLAD) technique [6] on a patterned surface so that

nanocolumns start to grow selectively on the surface and at a predetermined lateral distance. Similar to the case of Au spheres, the optimum distance here is also in the microns range. Vertical AuNRs can be prepared with nanolithography and selective etching processes relatively easy.

3.2 ESP-LSP coupling in gratings configuration

Grating coupling of ESPs can be obtained using variety of grating forms as mentioned before. The maximum field enhancement can be larger than the prism coupling case because of the sharp edges and corners of the grating lines. Figure 21 shows an EOT configuration slightly different from the one presented in Figure 11 so that the EOT peak is obtained at 1474 nm corresponding to the ESP generated at the top surface of the nanograting. The difference between the enhancement at the resonance or off resonance is small at the corners but significant on top of the lines, as one might expect since the corner-induced enhancement is more related to LSP excitation. When

Figure 21: Field distribution under EOT with and without metal NPs.

(A) Schematic of the nanograting on the substrate used for the EOT simulation. (B–C) Field distribution off resonance and on resonance. (D–E) Field distribution with three Au spheres on top of the grating lines at off-resonance and resonance wavelengths.

AuNPs are added on top of the grating lines, the ESP field couples to the LSPs, generating new hot spots, and the difference between the two cases of resonant and off-resonant coupling shows a 1 order of magnitude increase at the hot spots. Spreading NPs in a periodic manner and to be located at the exact position as drawn in the figure is not an easy task experimentally; however, with lithography, it is possible. Spreading NPs randomly with controlled average distance can be done easily and should reveal to comparable enhancement factors.

In a similar manner to the EOT case, Figure 22 presents field distribution calculations for AuNPs on top of thin dielectric grating on thin metal film configuration, with slight modifications from the case presented in Figure 10. The difference between the off- and at-resonance cases is

prominent. Interestingly, without the NPs, the field is concentrated at the edges of the dielectric grating lines, and when two NPs are located at these edges, the enhancement increased by an order of magnitude both at the top (Figure 11D–E) and the bottom (Figure 11F–G) corners. The concentration of the field at the Si_3N_4 edges indicates the formation of a standing wave with the line width equals to half the effective wavelength; that is, each Si_3N_4 line acts as nanoantenna along its width. The theory of plasmonic nanoantenna was formulated for the case of metal NPs [86–91]; however, here, we have a dielectric line on top of the metal film. The effective wavelength should satisfy $\lambda/2n_{\text{eff}} \approx 500 \text{ nm}$, because the distance between the center of masses of the field hot spots is nearly 500 nm, giving $n_{\text{eff}} = 1.527$, which is far from that for Si_3N_4 , but

Figure 22: Ultrahigh field enhancement under ESP-LSP coupling in thin dielectric grating and thin metal film geometry. (A) Thin dielectric grating on thin metal film scheme. (B–C) Field distributions calculated at and off resonance wavelength without NPs. (D–G) Field distributions calculated with NPs on the top and bottom corners at and off resonance wavelength.

simple homogenization for subwavelength grating reveals $n_{\text{eff}} \approx 1.56$. The difference might be due to the effect of the plasmonic film.

The ultrahigh enhancement achieved so far is at narrow wavelengths range. However, for many applications such as energy harvesting from the sun, for optoelectronic detectors efficiency enhancement, and for infrared spectroscopy, it is desirable to have ultrahigh enhancement over a wide spectral range. One approach to achieve this was done using the coaxial nanoantenna structure [27]; however, the field enhancement was nearly 300. To increase it further, we propose using the ESP coupling with the LSPs generated by the coaxial nanoantenna, for example by exciting it via prism or grating coupling. Another proposed approach uses thick metal grating on thick metal. As it is well known, thick metal grating gives many plasmonic and cavity resonances [106], and one can tailor them to be in the spectral range of interest. Based on this concept, ultrahigh enhancement was achieved over a wide spectral range and particularly by spreading NPs over the grating area. Figure 23 shows the case of 180 nm -thick Ag grating on a 100 nm -thick Ag film, that the enhancement is around 1000 for the whole visible range and around 2000 for the SWIR range. At 684 nm , a clear ESP mode is observed, while at 1748 nm , a clear cavity mode is observed, in which most of the energy exists in the

space between two neighboring grating lines. At the other wavelengths (538 nm and 1235 nm), a mixture of SPR and cavity modes are obtained.

As NPs are added on top of the grating lines, the ESP-LSP and cavity mode-LSP coupling reveals a higher field enhancement by 2 orders of magnitude. Figure 24 shows the field distribution calculations when three AuNPs are arranged on the edge/center/edge configuration. Note the strong effect of the spacer layer in Figure 24. It shows that an improvement of at least 1 order of magnitude is obtained in the enhancement factor at the spectral range between 530 and 950 nm when 2 nm spacer is used instead of 4 nm due to the stronger ESP-LSP and cavity-LSP mode couplings. Note that when using 2 nm instead of 4 nm spacer thickness, the peak at 844 nm shifted to 870 nm , while those at 662 nm and 734 nm remain the same. The shift can be understood as a result of the effect of the surrounding dielectric medium on the LSP and ESP resonances.

To increase the field enhancement in the 1000 nm – 1400 nm region, one can make the grating even thicker and optimize the grating period so that additional resonances appear in this spectral range. One of the advantages of using grating geometry is the ability to design and build them in different configurations, heights, periods, double periods, and different materials stacked together.

Figure 23: Ultra-high field enhancement under ESP-LSP and cavity modes-LSP coupling in thick metal grating geometry. (A) Schematic of thick metal grating on thick metal film. (B) Reflection and transmission spectra without the NPs with the grating and the dielectric spacer thicknesses are 180 nm and 4 nm, respectively. The calculations were done under TM polarization. (C) Maximum field intensity enhancement versus wavelength. (D) Intensity distribution at some resonances observed in the reflection spectrum. The legends in the figures in (D) show the maximum intensity enhancement at the specific wavelengths. Reproduced with permission from [106].

Figure 24: Spacer thickness effect on the field enhancement under thick metal grating geometry.

(A) Maximum field intensity enhancement versus wavelength for the edge/center/edge NP grating configuration under TM polarization as in Figure 22A. The grating and spacer thicknesses are 180 nm and 2 or 4 nm, respectively. (B) Intensity distribution at different wavelengths. The legends in the figures in (B) show the maximum intensity enhancement at the specific wavelengths. Reproduced with permission from [106].

The polarization dependence exists also in the 1D grating in Figure 23A because the NPs are arranged along the grating lines as a 2D array; as it was shown in [106], some resonant peaks appeared with transverse electric (TE) polarization. One can also investigate 2D and 3D grating structures to obtain polarization independence. Angular dependence is another issue for energy harvesting from the sun, which can be resolved by designing gratings at multiple periods. The recent advances in nanofabrication techniques allow for manufacturing a variety of grating structures at large scale, which, together with these novel designs, should reveal a plethora of improved optoelectronic devices, energy-harvesting methodologies, and surface-enhanced, spectroscopy-based sensors.

3.3 Other ESEW-LSP coupling configurations

The ultrahigh local field enhancement is not limited to ESP-LSP coupling, but other ESEWs can be used to excite LSPs and enhance the local field much higher than plane wave excitation. A variety of ESEWs exist other than

plasmonic waves, such as Zennek ESEWs, Bloch ESEWs, and Tamm ESEWs. The reader is referred to the modern book on SEWs [95]. Here, we give two examples of such waves generated at the surface of dielectric structures. The first what is called GMR that uses a single thick dielectric grating on top of substrate and the second is a 1D periodic stratified structure that is able to support Bloch ESEWs. In Figure 25, the GMR structure is shown with top arrays on AuNPs and light incident normally at TM polarization. This structure is known to resonate at certain wavelengths, giving resonant reflection at that particular wavelength. The thick grating is a subwavelength grating, so no diffracted light emerges. The basic principle relies on the fact that the structure can support guided modes at specific wavelength, which, upon constructive interference while bouncing back and forth in the waveguide, occurs in the backward direction. The usual way of obtaining it is by using thin dielectric grating on top of a thin waveguide layer, but here, the thick grating acts both as thin grating for coupling to the waveguide and as the waveguide layer itself. At resonance, the wave is evanescent and therefore confined at least in the direction perpendicular to the

Figure 25: Ultrahigh field enhancement under GMR-LSP coupling.

(A) Guided resonant grating structure with AuNPs on top. (B–C) Field distribution calculations without the AuNPs at and off resonance.

(D–G) Field distribution calculations with the AuNPs on the top or bottom corners at and off resonance.

grating surface similar to ESP wave. The resonance wavelength is determined by the product of the grating period and some effective refractive index of the mode $\lambda_r = n_{\text{eff}} \Lambda$, which in our case it is at 1469 nm when the grating is in water medium. The field distribution calculation shows 2 orders of magnitude enhancement between the two cases of off and at resonance (Figure 25). When the AuNPs are added on top, the field enhancement jumps to 2344 on the hot spots at the resonance, while it is only 190 without the AuNPs. Interestingly, as the AuNPs are located on the bottom corners, then the enhancement is slightly lower even though without the AuNPs, it was larger at the bottom corners than the top ones. Since the GMR structure is useful as a biosensor and tunable filter [107], now, we have a methodology to use it also for surface enhanced spectroscopy, energy harvesting, and optoelectronic devices.

Bloch SEWs (BSEW) are known to exist at the interface of a periodic structure and semi-infinite dielectric medium. The periodic structure can be one, two, or three dimensional, and usually, there is a need for some absorption; that is, the imaginary part of the dielectric function is nonzero. In stratified periodic structure, there is a need for the prism coupling in order to provide the k -vector matching to excite the BSEWs, as shown in Figure 26A. The BSEW excitation causes a dip in the reflectivity, as shown

in Figure 26B. When the AgNPs are added, there is a shift in the reflection dip. The field enhancement calculation shows improvement by more than an order of magnitude when the AuNPs are added. This enhancement is determined by the polarization used, the size of the NP, and the wavelength. It follows the main concept of ESEW-LSP ultrahigh enhancement mechanism, but it can be optimized further.

4 Summary and future perspectives

Ultrahigh enhancement of EM local fields obtained by exciting LSPs via ESPs and, in general, via any ESEW is a promising methodology for improving the efficiency of optoelectronic devices, sensing, and molecular research with surface-enhanced spectroscopies, solar energy harvesting, enhanced photocatalysis, and many other known plasmonic assisted processes. It can be done via prism, grating, waveguide, fiber, or other forms of coupling. The enhancement factors can be as high as 10^6 and more, where in using standard LSP excitation techniques, the best enhancement is lower by more than an order of magnitude. Thinking of optical emission signals from molecules that are proportional to $|E|^2$ (SEF and SEIRA)

Figure 26: Bloch ESEW coupling configuration with LSPs (TE polarization).

(A) Schematic. (B) Reflectivity of the structure. (C) Field amplitude enhancement factor. Parameters: $n_L = 1.45 + i0.0002$; $d_L = 247$ nm; $n_H = 2.096 + i0.0002$; $d_H = 130$ nm; $n_p = 1.518$; $n_s = 1.33$; $\lambda = 543$ nm; AgNP with 20 nm radius.

or $|E|^4$ (SERS), the emission efficiency can be enhanced significantly, thus boosting the possibility of detecting single molecules easier, superresolution imaging, and biosensing with ultralow detection limits using low power sources. These spectroscopic techniques provide another important sensing parameter, which is specificity, particularly Raman and infrared spectroscopies.

The manufacturability of structures giving ultrahigh field enhancement at large scale and in a cost-effective way needs to be progressed more, although at small scale, the methods exist, but for mass production, more needs to be done. Advances in synthetic and lithographic fabrication techniques allow researchers to tune the localized resonance wavelength through the visible, near infrared, and into the mid and long wave infrared regions of the EM spectrum, by varying the shape, size, and material of the NPs that support the LSPR [108]. Similarly, ESEWs and extended SPR (ESPR) can be generated almost in the whole spectral range using materials other than noble metals such as oxides and nitrides for the infrared range or using periodic structures of arrays of particles, gratings,

and photonic crystals. Stacked arrays such as *metal-insulator-metal* and *insulator-metal-insulator* structures are known to reveal long-range plasmons [109, 110]. Manufacturing techniques of nanostructures in large scale are being developed in the nanoelectronics industry and soon are expected to be available for nanophotonic applications as well at low cost. For example, academic institutions are now purchasing fast electron beam lithography machines and UV laser interference lithography machine. Deposition techniques of nanostructures in thin film form are another methodology; for example, sculptured thin films (STFs) are nanostructured inorganic materials with anisotropic properties that are prepared by the oblique angle deposition technique [111]. They show the potential to be designed in a controllable manner using physical vapor deposition methods for potentially multifunctional materials and products. The GLAD technique is a sophisticated method used to create 3D chiral nanostructures with a tailored geometry. Here, the self-organized nanostructure growth is based on a concurrent growth mechanism due to geometrical shadowing in combination with

kinetic limitation for surface adatoms. This technique requires a particle flux reaching the substrate under an extremely oblique angle of incidence. The growth conditions support a columnar growth, and the samples consist of 3D needles, which are slanted into the direction of the particle flux. As compared to existing methodologies of preparing metallic nanoparticles and porous silicon, STFs exhibit a wider range of possibilities, yet they cover the benefits that one expects from such biosensing systems. The porosity can be engineered to be within the 10–90% range. Recent research of metallic nano sculptured thin films (nSTFs) for plasmonic sensing showed great promise, particularly as SERS substrates. As these substrates can be patterned to control the distance between the nanocolumns, one can think of a scheme with ultrahigh SERS enhancement using the slanted nanorods on top of the closed metal film in the prism coupling configuration described in Figure 20. Metallic nSTFs can be classified as plasmonic metamaterials with unique properties having enhanced dielectric function. It is a plasmonic metamaterial operating at optical frequencies, another emerging group of materials with potential for ultrahigh field enhancements. The close-packing of metallic nanostructures is another approach that can boost the local electric field over the large area, in that overall epsilon of the optical effective medium can be unnaturally increased [112–116]. The close packing of plasmonic NPs into a ring inclusion was found to contain unnatural magnetism [117]. In particular, a strong electric field confinement via capacitive coupling between plasmonic NPs can drive a circulating displacement current, so as to make a negative refractive index at optical frequency more realistic. Hence, local field enhancement via plasmonics can greatly extend the accessible range of both electric permittivity and magnetic permeability beyond naturally occurring limits [118, 119]. Another fabrication methodology of interest is using bio-inspiration and particularly when making structures made of soft materials in what is called soft photonics, which allows making tunable plasmonic properties [120].

Together with the advancement in these nanofabrication and novel metamaterial design techniques, the new methodology for obtaining ultrahigh field enhancement is expected to provide a new family of devices. As the phenomenon has a multidisciplinary nature, it opens a new niche of research in metamaterials and nanophotonics with many innovative works expected to originate. Specifically, more works are required for (i) deep understanding of the origin of ultrahigh enhancement compared to the existing structures; (ii) demonstrating its existence with other ESP configurations, such as waveguides, fibers, 2D gratings, long-range plasmons, as well as coupling with other

types of ESEWs; (iii) finding the optimum conditions of the combination of parameters: coupling method, NP size, shape and separation versus the excitation wavelength for SEF, SERS, and SEIRA, as well as extending the spectral and angular excitation ranges to cover as much as possible of the solar spectrum over a wide angular range; and (iv) implementing the methodology to build devices with high performance such as sensors with high specificity and sensitivity, superresolution imaging, improved smart windows, optoelectronic devices with high efficiency, enhanced solar photocatalysis systems, smart windows improvement, and solar water heating and evaporation.

Due to its technological and scientific importance, ultrahigh sensitivity has a multiple of impacts on society. The science behind the ultrahigh optical field enhancement that occurs at the nanoscale neighborhood of metallic nanostructures and understanding its origin with new structures and excitation schemes will certainly impact the research of these systems by other interdisciplinary experimental and theoretical researchers. Light-matter interaction at strong optical fields is a hot topic of research, and the methodology proposed here will help researchers in the field to perform experiments using relatively more compact and low-cost optical equipment. The development and optimization of this new methodology will positively impact the efficiency and enhance the performance of photonic and energy-harvesting devices. Fabrication of novel and miniature devices that so far was not feasible with the existing technology will now be possible using the proposed nanostructures and photonic schemes. These include improved efficiency semiconductor photovoltaics, improved detectors for the infrared range, biosensors and environmental sensors with ultralow detection limit, and more improved solar energy harvesting systems such as using photocatalysis and sea water desalination. The technological advances likely to occur during the course of the coming decade will help spawn new companies and assist existing companies to remain competitive and profitable.

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