Remote grating-assisted excitation of narrow-band surface plasmons

Tae-Woo Lee1,* and Stephen K. Gray2

1Center for Computation and Technology, Louisiana State University, 216 Johnston Hall, Baton Rouge, Louisiana 70803, USA
2Center for Nanoscale Materials, Argonne National Laboratory, Argonne, Illinois 60439, USA
*twelee@cct.lsu.edu

Abstract: We show, based on theoretical analysis and realistic simulations, how a grating embedded in a dielectric substrate can excite surface plasmon polaritons (SPPs) on the top side of a flat metal film far removed from the grating. This remote SPP excitation is characterized by a narrow spectral bandwidth and a high near-field intensity relative to the standard approach for exciting SPPs. The simplicity of the structure and the fact that it requires only normally incident light should make it relevant to the many applications that benefit from high quality SPPs on a flat metal film.

©2010 Optical Society of America

OCIS codes: (240.6680) Surface plasmons; (120.2230) Fabry-Perot; (130.6010) Sensors; (000.4430) Numerical approximation and analysis.

References and links

1. C. K. Chen, A. R. B. de Castro, and Y. R. Shen, “Surface-enhanced second-harmonic generation,” Phys. Rev. Lett. 46(2), 145–148 (1981).
2. V. A. Markel, V. Shalaev, P. Zhang, W. Huyhn, L. Tay, T. Haslett, and M. Moskovits, “Near-field optical spectroscopy of individual surface-plasmon modes in colloid clusters,” Phys. Rev. B 59(16), 10903–10909 (1999).
3. Y. Hamanaka, K. Fukuta, A. Nakamura, L. M. Liz-Marzán, and P. Mulvaney, “Enhancement of third-order nonlinear optical susceptibilities in silica-capped Au nanoparticle films with very high concentrations,” Appl. Phys. Lett. 84(24), 4938 (2004).
4. J. P. Huang, and K. W. Yu, “Optical nonlinearity enhancement of graded metallic films,” Appl. Phys. Lett. 85(1), 94 (2004).
5. J. Toudert, H. Fernandez, D. Babonneau, S. Camelio, T. Girardeau, and J. Solis, “Linear and third-order nonlinear responses of multilayered Ag: Si,N nano-composites,” Nanotechnology 20(47), 475705 (2009).
6. K. Kneipp, Y. Wang, H. Kneipp, L. Perelman, I. Itzkan, R. Dasari, and M. Feld, “Single molecule detection using surface-enhanced Raman scattering (SERS),” Phys. Rev. Lett. 78(9), 1667–1670 (1997).
7. J. M. Montgomery, A. Imre, U. Welp, V. Vlasko-Vlasov, and S. K. Gray, “SERS enhancements via periodic arrays of gold nanoparticles on silver film structures,” Opt. Express 17(10), 8669–8675 (2009).
8. A. Baca, T. T. Truong, L. R. Cambrea, J. M. Montgomery, S. K. Gray, D. Abdula, T. R. Banks, J. Yao, R. G. Nuzzo, and J. A. Rogers, “Molded plasmonic crystals for detecting and spatially imaging surface bound species by surface-enhanced Raman scattering,” Appl. Phys. Lett. 94(24), 243109 (2009).
9. T. W. Ebbesen, H. J. Lezec, H. F. Ghaemi, T. Thio, and P. A. Wolff, “Extraordinary optical transmission through sub-wavelength hole arrays,” Nature 391(6668), 667–669 (1998).
10. S. H. Chang, S. K. Gray, and G. C. Schatz, “Surface plasmon generation and light transmission by isolated nanoholes and arrays of nanoholes in thin metal films,” Opt. Express 13(8), 3150–3165 (2005).
11. M. E. Stewart, N. H. Mack, V. Malyarchuk, J. A. Soares, T. W. Lee, S. K. Gray, R. G. Nuzzo, and J. A. Rogers, “Quantitative multispectral biosensing and 1D imaging using quasi-3D plasmonic crystals,” Proc. Natl. Acad. Sci. U.S.A. 103(46), 17143–17148 (2006).
12. H. Raether, Surface Plasmons on Smooth and Rough Surfaces and Gratings (Springer, Berlin, 1988).
13. K. A. Willets, and R. P. Van Duyne, “Localized surface plasmon resonance spectroscopy and sensing,” Annu. Rev. Phys. Chem. 58(1), 267–297 (2007).
14. T. W. Lee, and S. K. Gray, “Subwavelength light bending by metal slit structures,” Opt. Express 13(24), 9652–9659 (2005).
15. A. Taflove, and S. C. Hagness, Computational Electrodynamics: The Finite-Difference Time-Domain Method, 2nd ed. (Artech House, Boston, 2000).
16. T. W. Lee, and S. K. Gray, “Regenerated surface plasmon polaritons,” Appl. Phys. Lett. 86(14), 141105 (2005).
17. J. M. Montgomery, and S. K. Gray, “Enhancing surface plasmon polaritons: propagation lengths via coupling to asymmetric waveguide structures,” Phys. Rev. B 77(12), 125407 (2008).
18. M. Weisser, B. Menges, and S. Mitterle-Nehrer, “Refractive index and thickness determination of monolayers by multi mode waveguide coupled surface plasmons,” Sens. Actuators B Chem. 56(3), 189–197 (1999).
1. Introduction

Surface plasmons (SPs) are electromagnetic excitations induced by collective oscillations of free electrons at the interface between metals and dielectrics. The evanescent character of SPs, coupled with large near-field intensity enhancements, make them attractive for enhancing nonlinear optical processes [1–8], including surface enhanced Raman scattering (SERS) [6–8]. The extraordinary optical transmission (EOT) [9,10] of light that can occur through periodic slits or hole arrays in metal films is another SP-related phenomenon of relevance to refractive index sensing, for example [11].

While manifestations of the same basic phenomenon, one can distinguish between two types of SPs: surface plasmon polaritons (SPPs) on metal films which are traveling waves, or standing waves composed of counter propagating SPPs [12], and local surface plasmons (LSPs) which are associated with excitations of individual nanoparticles [13]. Our main concern is with SPPs, although the structure we present may be relevant as a component in hybrid systems that involve LSPs such as the metal nanoparticle/periodic slit system of Ref [7]. SPPs have wave vectors or momenta greater than ordinary light and are excited by transferring an appropriate amount of momentum into the system. This is also called phase matching. One excitation approach is attenuated total reflection (ATR) [12], in which a thin metal film is sandwiched by two different dielectric materials and light is incident with a specific angle relative to normal from the optically denser dielectric side so as to satisfy a phase matching condition. Another approach is the use of gratings which can provide additional phase matching from the generation of integer multiples of the reciprocal lattice vector [8]. Gratings can also be used with normally incident light, which is simpler from a practical standpoint. For example, normally incident light on periodic slits or hole arrays in metal films can lead to Bloch wave SPPs (BW-SPPs) of relevance to the EOT noted above [9,10]. However, the non-uniform surface features in these systems impose limitations. For example, the grating can add friction to the flow of an analyte when it is combined with micro-fluidic channels.

Here we introduce a simple system that can achieve high quality SPPs on a flat film surface with just normally incident light. Realistic, finite-difference time-domain (FDTD) calculations [14,15] and detailed analysis reveal that SPPs with a high near-field enhancement and an ultra-narrow bandwidth can still be excited with a grating located far away from the flat thin film. We believe that this approach for generating SPPs is fundamentally interesting and also could be useful in many applications, e.g., in improving SERS sensing capabilities without the need of narrow band tunable lasers or laser line filters to block undesired light from an input laser source. The flat surface also provides freedom to engrave additional patterns on the surface to achieve the better control of SPPs on that surface, e.g., bow-tie shapes.

2. System description and working principles

The system consists of a grating structure embedded in a dielectric region, a metal film on top of this region and an optically less dense region above (Fig. 1). Unlike conventional ATR, light is normally incident from below. The light interacts with the grating, producing a certain angle of diffracted light toward the top metal thin film such that
where $m$ is an integer representing a grating mode, $\lambda_0$ and $P$ are the wavelength of light and the grating period, respectively, and $n_2 = \sqrt{\varepsilon_2}$ is the refractive index in medium 2. With a semi-infinite metal approximation, the SPP condition on the top of the flat metal film interfacing with $\varepsilon_1$ is [16]

$$\sin \theta_{\text{SPP}} = \frac{\varepsilon_{\text{metal}}'(\lambda_0) \varepsilon_1}{\varepsilon_2 \left[ \varepsilon_{\text{metal}}'(\lambda_0) + \varepsilon_1 \right]},$$

(2)

where $\varepsilon_{\text{metal}}'(\lambda_0)$ is the real part of the wavelength-dependent dielectric constant of the metal. (For simplicity we assume the imaginary part $\varepsilon_{\text{metal}}'' << \varepsilon_{\text{metal}}'$.) SPP excitation is possible when the angles $\theta_{\text{GM}}$ and $\theta_{\text{SPP}}$ are equal, which results in

$$\frac{m \lambda_0}{P} = \frac{\varepsilon_{\text{metal}}'(\lambda_0) \varepsilon_1}{\varepsilon_{\text{metal}}'(\lambda_0) + \varepsilon_1}.$$ 

(3)

Interestingly, this equation is identical to the condition for a BW-SPP on the metal-$\varepsilon_1$ interface [10], and as such we are effectively exciting BW-SPPs on a flat, non-periodic surface. The physical excitation mechanism involved, however, is different from the BW-SPP case. In particular, Eq. (2) must apply and for it to yield a real angle for the typical case that $|\varepsilon_{\text{metal}}'|$ is much larger than $\varepsilon_1$, we find the additional condition that $\varepsilon_2 > \varepsilon_1$. This latter condition is not required for BW-SPP excitation. We call our proposed system a remote-grating, or r-grating system.

Fig. 1. Schematic illustration of the remote grating SPP setup. Dot-patterned areas denote metallic regions. Bottom grey and top white regions represent dielectrics with dielectric constants $\varepsilon_2$ and $\varepsilon_1$. Light is incident from below. The embedded grating processes the light to form diffracted light at angle $\theta_{\text{GM}}$. (Modes with angle $\theta_{\text{SPP}} - \theta_{\text{GM}}$ are also excited.) The system is infinitely extended in the lateral direction. SPPs on the top metal film are excited when the grating mode angle is matched with the SPP angle.

3. Numerical analysis and results

Considering a system layout in an x-y domain, and assuming z invariance, the problem is effectively reduced to a 2-D problem. 2-D FDTD simulations with $E_x$, $E_y$, and $H_z$ field components are carried out, with normally incident, TM polarized light. A Drude plus two-pole Lorentz model is used to define a realistic dielectric constant for gold [11], which is taken to compose both the grating and the thin film. An auxiliary differential equation technique [14,15] is used to incorporate this wavelength-dependent dielectric constant into the
calculations. The top and bottom dielectric regions are filled with \( \varepsilon_2 = 1 \) (air) and \( \varepsilon_2 = 2.3104 \) (glass), respectively. Periodic boundary conditions are applied on the left and right sides, and the top and bottom sides are truncated with uniaxial perfectly matched layers [15]. A time-windowed plane wave with frequency content in the range of interest is launched below the metal grating structure in the bottom region using the total-field/scattered-field approach [15]. Fourier transforms are used to generate the frequency (or wavelength) dependent fields. For all simulations, uniform 5 nm grid spacing is used in \( x \) and \( y \).

We first consider a 40 nm thin gold film with glass substrate and air above. Within the substrate, we have a grating of periodicity \( P = 800 \) nm composed of 200 nm × 100 nm gold bars. The distance between the bottom of the grating and the top of the gold film, \( h \) in Fig. 1, is 1050 nm. We have designed our particular illustration of the r-grating system to yield a resonance near \( \lambda_0 \approx 800 \) nm. This wavelength is relevant to Raman spectroscopy for biological cell samples. (Near this wavelength, background noise in a Raman signal from naturally occurring fluorescence scattering is suppressed.) Figure 2(a) shows maximum intensity enhancements, defined as \( |E|^2 \) measured just above (~10 nm) the top flat gold film and normalized by the field intensity of the incident light. Intensity distributions are shown in Fig. 2(b). Between the bottom grating and top metal film, interferences associated with a grating mode are observed. Due to the bidirectional propagations, intensity distributions of SPPs exhibit a standing wave pattern. The intensity maxima along \( x \) turn out to be at \( x_c \pm P/4 \), where \( x_c \) represents the midpoint of a grating bar down below in the glass. For comparison, SPP enhancements from an ATR configuration (glass-40 nm metal film-air) are calculated using the Fresnel equations and are also shown.

As shown in Fig. 2(a), the peak wavelength is near 811 nm. This is in reasonable accord with Eq. (3), which for \( m = 1 \) yields solution \( \lambda_0 = 815 \) nm and insertion of this value into either Eq. (1) or (2) yields \( \theta_{GM} = \theta_{SPP} = 42.1^\circ \). At peak maximum, the local field enhancement for r-grating reaches 245, while the ATR case is 120, a half of the r-grating enhancement. The r-grating system is supposed to suffer from reflection losses at the bottom grating and, thus weaker intensity light is delivered to the top metal film. However, radiation leakage from SPPs on the top metal film back to the bottom dielectric medium, can also be blocked by the bottom grating, redirecting radiation losses back towered the top metal film to re-generate SPPs. Hence, it produces a higher SPP intensity that compensates the reflection losses at the bottom grating. This mechanism is similar in spirit to previously studied R-SPP system [16,17]. The intensity enhancement spectrum of the r-grating in Fig. 2(a) shows a very narrow
peak width. The full-width of half-maximum (FWHM) is measured as 3 nm. Compared to a SPP peak generated from ATR setup, with FWHM ~110 nm, the r-grating SPP peak is 36 times narrower. If this system is applied in a SERS context, for example, the ultra-narrow SPP band can serve as having an effective laser line filter with a 3 nm window in the laser input module. We note that thicker top metal films produce even narrower FWHM, but the SPP intensity decreases. (The minor peak structure to the longer wavelength of the main resonance in Fig. 2(a) can be attributed to a state that has less SPP character and more waveguide or FP mode character.) This comparison study with the well-established ATR-SPP case gives a general idea of how the r-SPP performs. It also provides a reference to be compared with other SPP excitation techniques such as waveguide-coupled surface plasmon resonances (WCSPRs) [18].

During SPP generation, the first light interactions occur at the grating. The width of each metal bar of the grating (oriented as in Fig. 1), denoted $w$, influences system performance. For smaller $w$, initial reflection losses from the grating are reduced such that a larger portion of incident light passes to the top metal film. Meanwhile, it reduces the SPP regeneration mentioned above, i.e., blocks radiation loss. Conversely, larger $w$ increases the initial reflection losses but helps favorable SPP regeneration. Therefore, an optimum $w$ exists. To examine this, we gradually increase $w$ from 120 nm to 450 nm with 10 nm steps while fixing every other structural parameter with the same values for the case of Fig. 2. The results are presented in Fig. 3. Intensity enhancements start from 49.4 at $w = 120$ nm and reach to peak value, 245 at $w = 200$ nm. Then, the enhancement gradually decreases for larger $w$. The FWHM is largest at the smallest $w$ (120 nm), sharply decreases for larger $w$ until 200 nm, and then decreases more slowly. At $w = 200$ nm, the FWHM is still very close to its smallest value and is $\approx$3 nm, thus motivating our original system design. In contrast, the height of the metal bar has a small effect on the overall results. We verified this by varying the thickness of the metal bar from 70 nm ~110 nm, with the corresponding enhancements ranging from 255 to 235.

![Fig. 3. Peak intensity enhancement and FWHM of SPP peaks as a function of the width of grating metal bar, $w$. A blue solid line with open circle marks and a green solid line with asterisk marks represent maximum intensity enhancement and FWHM at each $w$, respectively.](image)

In addition to diffracting light in order to generate phase-matched SPPs on the top film, light refracted down from the top film can be reflected back up by the grating. The grating-glass-film can thus form a Fabry-Pérot (FP) resonator. Hence, the distance between the bottom grating layer and top metal film, denoted $h$, also becomes a determining factor for SPP properties. We conduct FDTD computations for a variety of $h$ values. The particular case $h = 0$ is similar to a conventional BW-SPP system, i.e., the grating metal bars are attached to top metal. The $h$ values are extended up to 2430 nm with 20 nm increments. The other structural parameters are kept same as in Fig. 2, including $w = 200$ nm. The intensity enhancement plot, Fig. 4(a), shows several minima and maxima as $h$ is varied. Some maxima exhibit sharp peaks while others are rather broad. In design considerations for obtaining high quality SPPs, the system provides several $h$ windows that produce relatively high enhancements. For example,
windows of 270 – 410 nm, 1040 – 1150 nm, 1330 – 1470 nm, … produce enhancements greater than 120. Overall, the size of the h window for such enhancements is about 100 nm. The minima exhibit a periodic pattern that is repeated roughly in every \( \Delta h_{\text{min}} \approx 270 \) nm. The SPP peak wavelength, not shown here, varies only slightly with \( h \), in the 805 – 815 nm range.

The pattern of minima and maxima in Fig. 4(a) can be qualitatively understood using the well-known expressions [19] for the positions of the FP modes between two identical flat films, with \( j = 0, 1, 2 \):

\[
\frac{4\pi n_h\cos \theta_{\text{FP}}}{\lambda_0} = (2j+1)\pi.
\]  

(4)

where the even case corresponds to transmission maxima and the odd case to transmission minima (or reflection maxima). We term these cases “resonant” and “anti-resonant” FPs, respectively. The zero-order FP modes correspond to \( \theta_{\text{FP}} = 0 \). The observed nearest spacings between minima in \( h \) are \( \Delta h_{\text{min}} \approx 270 \) nm [Fig. 4(a)]. This correlates well with \( \lambda_0 / (2n_h) \approx 268 \) nm (using \( \lambda_0 = 815 \) nm) which would be expected from Eq. (4). It seems very plausible that the anti-resonant FP modes, which represent reflection maxima, lead to intensity minima on the upper surface since only a small amount of light will reach it. However, the specific minimum positions in Fig. 4(a) can deviate from those predicted by Eq. (4), \( h = (2j+1)\lambda_0 / (4n_h) \), by up to \( \approx 100 \) nm. This is due to additional non-zero phase shifts on reflections at the top metal film and the bottom metal grating [20] which are not taken into account in Eq. (4).

The origins of the intensity maxima pattern in Fig. 4(a) are also interesting. One might naively correlate them with the approximate positions of the resonant zero-order FP modes which correspond to transmission maxima. However, since all zero-order FP modes have \( \theta_{\text{FP}} = 0 \), they cannot provide direct phase matching into the SPPs on the upper surface. Instead, first order FP modes, with \( \theta_{\text{FP}} \approx 42.1^\circ \) in Eq. (4), can provide the necessary coupling. This situation therefore corresponds to \( \theta_{\text{GM}} = \theta_{\text{FP}} = \theta_{\text{SPP}} \). These modes can be directly excited or they could be initiated from the zero-order resonant FP modes which are bouncing back and forth between the grating and the film, storing energy in the FP cavity. This interpretation is consistent with most of the maxima observed in Fig. 4(a). However, there are two maxima, one at \( h = 1050 \) nm and another at \( h = 2130 \) nm, that are noticeably larger than the others.

![Fig. 4. Intensity enhancement, (a), and FWHM, (b), of SPP peaks as a function of the distance between grating metal layer and top metal film, h.](image-url)

Interestingly, in both these cases there is a very close intensity minimum. This minimum correlates with an anti-resonant, zero-order FP mode that is highly reflecting, allowing the first-order modes to couple directly with the film. Since SPP generation processes should be more efficient with direct interactions from the first-order grating mode, one obtains the highest SPP intensities in these two peak structures. The FWHM in Fig. 4(b) shows only small deviations around the two major peaks, just 3 – 4 nm, consistent with a very high quality resonance excitation process occurring in those two spectral regions. We note that for \( h = 0 \),
the structure no longer provides a remote grating effect. Rather, it corresponds to a conventional BW-SPP type grating. In Fig. 4(a), the enhancement curve undergoes a sharp variation and turnover for decreasing $h$ below 50 nm. For $h$ ranging from 50 nm to 10 nm, the enhancement drops from 151 to 30. However, upon decreasing $h$ further a turnover point is reached with enhancement increasing until the conventional BW-SPP case, enhancement 80, is achieved at $h = 0$. The scale of Fig. 4(a) is too large to see this turn over. See, however, Fig. 5 that illustrates the intensity distributions in this region. This rapid change of enhancement with small variation in $h$ is due to local near field interactions between the grating and top metal film. As $h$ approaches zero, the system forms a narrow gap between grating layer and top metal film. In this case, local resonances occur, concentrating energy in this region. The energy contributing to the top SPPs is then reduced [Fig. 5(b)], decreasing top SPP intensities. When the bottom metal grating and top film are attached each other, the gap disappears and, thus, the SPP intensity is increased to the BW-SPP value.

![Fig. 5. Transition from an r-grating to a conventional BW-SPP type grating. Intensity distributions over one grating period with different $h$, (a) 50 nm, (b) 10 nm, and (c) 0 nm are shown. Highly localized fields between grating bar and metal film are visible in $h = 10$ nm case with decreased SPP intensity. White lines denote metal structures. (In (b), metal structures are not outlined in order to show clearly the localized intensity between the top of the grating and the bottom of the film.)](image)

We should stress, though, that remote SPP excitation is not necessarily superior to other means of exciting SPPs. For example, by varying the width of the grating one could probably improve the $h = 0$ conventional grating enhancements but of course the remote feature is lost. See also other recently suggested structures for SPP generation from various slit structures and on periodically patterned metal films [21–23]. We wish to put forth remote SPP excitation as simply a new, and we believe interesting, means of exciting high quality SPPs on a flat metal film that could offer advantages for some applications.

As we discussed, the grating layer provides an intermediate step to support phase matching light interactions to SPPs on a remotely located flat metal film. Other forms of gratings could lead to the same remote SPP effect. For example, a non-metallic grating with strong dielectric contrast could be used as an alternative. Additional flexibility could be provided by use of an addressable liquid crystal layer to form the grating.

4. Summary and conclusions

In this study, we presented a new approach to excite SPPs on a flat metal film without restrictions of the incident illumination angle. The system consists of a top metal film on a dielectric in which a periodic grating structure is embedded. Through diffraction grating modes, SPP excitation becomes possible with normally incident light which makes the system appealing from an experimental point of view. We also showed how additional enhancements can be provided by coupling to Fabry-Pérot modes. All these features lead to a reduction in
losses that are sometimes encountered with other ways of exciting SPPs. For example, the Fabry-Pérot aspect reduces leakage loss and the fact that SPPs are being generated on a flat film prevents scattering losses that would occur on structured films. As a consequence, our numerically rigorous FDTD analysis showed that high intensity SPPs with a very narrow band features can be generated in the structure.

The narrow band SPP response can provide advantages to SERS applications, as well as surface plasmon resonance (SPR) sensing. This system can be extended further to produce other desired outcomes specific to a certain application by modifying the grating layer. For example, multiple grating layers with several periodicities could be used for multiple wavelength SPP resonances. The r-remote system could also serve as a component in SPP/LSP hybridized SERS systems [7].

Acknowledgements

T.-W. Lee was supported by the National Institutes of Health (NIH) program under ARRA, grant No. 2R01EB004761-06. Computing resources were provided by Louisiana Optical Initiative Network, or LONI. Use of the Center for Nanoscale Materials was supported by the U. S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.