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Ultrasmooth metallic films with buried nanostructures for backside reflection-mode plasmonic biosensing

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Abstract

We present a new plasmonic device architecture based on ultrasmooth metallic surfaces with buried plasmonic nanostructures. Using template-stripping techniques, ultrathin gold films with less than 5 Å surface roughness are optically coupled to an arbitrary arrangement of buried metallic gratings, rings, and nanodots. As a prototypical example, we present linear plasmonic gratings buried under an ultrasmooth 20 nm thick gold surface for biosensing. The optical illumination and collection are completely decoupled from the microfluidic delivery of liquid samples due to the backside, reflection-mode geometry. This allows for sensing with opaque or highly scattering liquids. With the buried nanostructure design, we maintain high sensitivity and decoupled backside (reflective) optical access as with traditional prism-based surface plasmon resonance (SPR) sensors. In addition, we also gain the benefits offered by nanoplasmonic sensors such as spectral tunability and high-resolution, wide-field SPR imaging with normal-incidence epi-illumination that is simple to construct and align. Beyond sensing, our buried plasmonic nanostructures with ultrasmooth metallic surfaces can benefit nanophotonic waveguides, surface-enhanced spectroscopy, nanolithography, and optical trapping.

Keywords

Surface Plasmon Resonance (SPR); Biosensing; Template Stripping; Plasmonics; Nanostructures; Grating Couplers; Atomically Flat Gold Film; Nanolithography

Introduction

The field of plasmonics has undergone rapid development in the past decade due in part to its many promising applications for biosensing [1–4], imaging [5], photovoltaics [6], surface-enhanced spectroscopy [7–9] and data storage [10] among others. Surface plasmons [11] (SPs) are free electron oscillations that can be excited at optical frequencies in metallic films such as gold or silver. Using SPs in various metallic films, gratings, and nanostructures, biosensing in particular has seen significant progress [3,4,12–14]. Nanoholes [1,2,15–22], nanoparticles [23,24], and plasmonic interferometers [25,26] have all shown promise for enhancing the sensitivity, multiplexing capacity, and biological interfacing to augment the capability of conventional surface plasmon resonance (SPR) instruments. However, the simplest structure, a ~50 nm thick gold film on a prism for Kretschmann excitation of SPs, still remains the most widely used sensor setup [3,27–29]. For prism-

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based SPR detection, there is no need for nanofabrication of sensor chips and the reflective-mode geometry decouples optical paths in the prism from fluidic channels. If the optical path passes through the sensing fluid, as is the case with many transmission-mode nanoplasmonic biosensors [1,2] or grating-based SPR sensors [12,30], optical interference may occur, and the use of opaque, turbid, bubbly, or highly scattering fluids (such as blood or milk) is problematic. Unfortunately, the prism-coupling method also has several disadvantages: (1) Aligning and focusing the light source, imaging optics and detector at a large angle of incidence is cumbersome; (2) The sensing surface is tilted with respect to the optical axis, leading to image distortion. Furthermore, the depth-of-focus of the imaging lens limits the available sensing area which is problematic for high-resolution wide-field imaging of dense microarrays [16]; and (3) It is not trivial to tune the spectral resonances as it can require changing the illumination angle. For these reasons, many researchers have been exploring the use of nanoplasmonic structures that allow high-resolution, multiplex sensing at normal incidence with the use of standard optical microscopes, and offer wide ranges of resonant tuning by simply changing the geometry or materials [13]. These plasmonic sensing methods are summarized in Figure 1a–c. Other challenges remain, however, particularly in reproducible fabrication, performance, and smoothness of the metallic surfaces. Low-cost manufacturing of high-quality samples also presents a significant hurdle to the widespread adoption of nanoplasmonic sensors in research and commercial environments. In particular, surface roughness can lead to many detrimental effects, such as unwanted broadening and attenuation of plasmon resonances, sample-to-sample variation, reduced SP propagation lengths [31], and can also degrade the quality of self-assembled monolayers or supported lipid membranes formed on the metals [32,33]. Template stripping can alleviate many of these challenges by producing ultrasmooth unpatterned [34–36] or patterned [31,37–42] metallic surfaces with improved surface chemistry [43]. However, a device architecture for high-resolution SPR imaging, decoupled optical and fluidic access (i.e. backside illumination), and with integrated patterning and ultrasmooth metallic surfaces has not yet been demonstrated.

In this paper, we present a new device architecture that uses plasmonic nanostructures buried beneath a thin, ultrasmooth metallic top layer (Figure 1d). By leveraging the smooth films produced via template stripping and the design freedom of backside patterning, these devices retain some key advantages of conventional prism-based SPR instruments—namely a flat sensing surface with a reflection-mode geometry—while offering other advantages of nanoplasmonic biosensors—namely the wide optical tunability, normally incident light, and high-resolution imaging and multiplexing. Additionally, these buried plasmonic nanostructures could also be used for building nanophotonic circuitry and waveguides [44], spatial light modulators, optical nanoscopy, interferometric SP sensors [25], plasmonic traps [45], and present ultrasmooth sensing surfaces for self-assembled monolayers or lipid bilayers [46].

Methods

Figure 2 outlines the fabrication process of a buried SPR sensing device. A 20 nm thick layer of Au is first deposited by electron-beam evaporation on a Si wafer (Figure 2a). Next, electron-beam lithography followed by a 50 nm thick evaporation of Au and a lift-off process is used to define a linear grating of 50 nm tall Au stripes (Figure 2b) on top of the original film. The wet processing involved in our lift-off process leaves the fragile 20 nm thick Au film intact and attached to the Si template. After applying an adhesive backing layer (Norland 61 UV curable epoxy), the patterned device can be stripped from the Si template (Figures 2c,d). Critically, the patterns are fully buried in the backing layer and the top-side—previously in contact with the Si—is revealed to have an ultrasmooth surface, less

than 5 Å root-mean-square (RMS) roughness as measured by the atomic force microscopy (AFM) scans shown in Figure 3.

To demonstrate the versatility of our fabrication technique and to study the optical response of the buried grating structures, we fabricated a large array of gratings, varying both the period and linewidth. Figure 4a shows an optical micrograph (5x, 0.15 NA) with a 633 nm laser-line filter demonstrating the variations in reflection intensity as a function of line width and periodicity. Figures 4b,c show scanning electron micrographs (SEM) of a grating before and after template stripping, respectively. The surface after template stripping has a 4.8 Å surface roughness, and the buried grating is seen through the flat Au layer by using a 15 keV SEM accelerating voltage. Beyond gratings, the fabrication process described here can also be used to make a variety of other plasmonic nanostructures such as an embedded bull's eye [47] and an array of nanodots as shown in Figures 4d and 4e, respectively. Since the optical access only occurs in the half-space below the ultrasmooth top surface, the top surface is free to access fluids for biosensing applications. In our backside epi-illumination geometry, the light does not pass through the sample, allowing the use of complex, multi-layer microfluidic chips [48] and opaque liquids without the difficulties associated with aberration, absorption or scattering, which are problematic for grating-based SPR sensors [12] or transmission-mode nanoplasmonic sensors. Furthermore, since a single objective lens is used for both illumination and imaging at normal incidence, it is straightforward to align the optics and switch the imaging magnification or illumination wavelength.

Characterization of the devices consisted of Finite-Difference Time-Domain (FDTD) computer simulations, optical reflection spectral measurements, atomic layer deposition (ALD) layer-by-layer sensor response, and refractive index sensing with glycerol in water mixtures and colored or opaque liquids. FDTD simulations were run in 2D with a transverse-magnetic polarized (x -axis) incident pulse (directed along the $+z$ axis) that was Fourier transformed upon reflection to extract a full spectrum with a single simulation. The grid sizes were 1 nm in the x direction and 10 nm graded down to 1 nm in the z direction. Field penetration depths were determined by measuring the z -component of the electric field amplitude as it decayed to the “1/e” point. Optical reflectivity measurements were performed with an inverted Nikon microscope (Eclipse Ti) and a 5x objective (NA = 0.15) with a linear polarizer. For illumination, the back aperture of the objective was under-filled to provide near-normal incidence. The spectra were collected with an imaging spectrometer (Newport MS-257i). Liquid sensing experiments were performed with varying amounts of glycerol in water. The index of refraction was measured with a refractometer (Refracto 30PX, Mettler Toledo). Layer-by-layer ALD was performed by measuring a single device over many consecutive ALD runs. The reflection spectra were measured with the sensing side of the device immersed in water to obtain an accurate measure of the sensitivity in an aqueous environment [49]. The device was then dried with high-purity N_2 , the next layer of Al_2O_3 was added, and the same measurement procedure was performed. All reflection spectra were normalized to the reflection from an unpatterned area of the thin gold film.

Results and Discussion

The backside reflection spectrum measured in water from a Au grating device with a period of 500 nm and a line width of 200 nm is plotted in Figure 5a and shows three clear reflection dips. To understand the origin of these resonances, we performed FDTD simulations for the same device (Figure 5b). The 20 nm thick Au film is aligned at $z = 0$ nm, with the buried gratings for $z < 0$ nm. Water (refractive index, $n = 1.33$) fills the half-space with $z > 20$ nm and optical epoxy ($n = 1.56$) for $z < 0$ nm. The same three reflection dips were present in the FDTD simulations.

The origin of the different resonances can be attributed to the two-fold role of the bumps buried under the thin Au film. Teperik et al. observed a similar strong plasmon coupling behavior in a buried nano-void device [50]. In our device, the periodic bumps act as coupling elements to excite propagating SPs on the bump surface. Since the Au layer is ultrathin, these SPs also couple to the top gold-water interface. The light re-radiated from those coupled plasmons interferes with the directly reflected light and generates resonance dips in reflection. At visible frequencies, the penetration depth of the propagating SP modes into a surrounding dielectric medium is usually 100–300 nm [51] and in our case is 140 nm into the water. The second plasmon mode is the localized mode excited due to the individual bumps, which has a shorter lifetime—i.e. a broader resonance—than the propagating mode [52]. The field is mainly concentrated at the bump-substrate interface (here extending 30 nm from the bump into the substrate) and can easily couple back to light. The three reflection dips shown in Figure 5b were further investigated by examining the z-component of the electric field at each of the resonances (Figure 5c). The first resonance at 671 nm shows strong field confinement around the buried bumps, but also extends far from the top surface into the water (“water-side” grating resonance). The last resonance at 877 nm, on the other hand, shows a strong field extending from the buried grating on the substrate side (“substrate-side” grating resonance). The middle resonance (“coupled mode”) at 752 nm shows a combination of propagating and localized modes, with field penetration into the water side and field localization on the substrate side [50,53]. However, it also shows the shortest field penetration (80 nm) into the sensing (water) side of the Au film.

The trends of the reflection resonances are further investigated by looking at how these spectra vary with grating period both experimentally (Figure 5d) and in simulation (Figure 5e). For grating periods of 300 nm to 400 nm, the reflection dips at ~780 nm (simulation) and ~670 nm (experiment) are relatively insensitive to changes in period, indicating a localized resonance. This localized feature extends through all the grating periods examined (300 nm to 1000 nm), and is indicated by a dashed ellipse in the period vs. wavelength reflection colormaps. Due to the coupling of the grating plasmon modes with the localized plasmon modes, there are several peaks and dips throughout the localized band. As for the propagating modes, for a thin film, the plasmons on the two sides of the film will couple into a single insulator-metal-insulator (IMI) plasmon [54]. With an asymmetric structure (epoxy-gold-water) several modes will emerge. In our case, the IMI plasmon modes excited by a grating at normal incidence will have a wave number k_{spp} equal to that of the grating, i.e. $k_{spp} = 2\pi/a$, where a is the period. Solving the full asymmetric IMI dispersion relation as given by Burke et al. gives two grating-type IMI plasmons, plotted as the straight dashed lines over the reflection colormaps. The same lines are plotted over both the experimental and the simulated data. They are seen to shift significantly with period, indicating a grating-type plasmon. The dashed lines are also seen to intersect with the vertical localized plasmon band (dashed rectangle), causing peaks and dips to appear in the colormaps. These features allow us to conclude that the grating-type IMI plasmons and the localized effect of the grating bumps couple to form the various observed modes of our structures [55].

For sensing applications described herein, we chose the spectral feature that showed the best sensitivity to refractive index changes on the water-side. FDTD simulations showed that the “water-side” grating reflection dip at 671 nm shifted 300 nm per refractive index unit (RIU) change and that the “coupled resonance” at 752 nm shifted 240 nm/RIU. The peak between these two resonances at 720 nm shifted 356 nm/RIU and was therefore tracked for all sensing experiments. In previous work, spectral dips and peaks have also been shown to exhibit different spectral responses due to the excitation of grating-type or localized plasmon modes [56]. The localized resonance of the individual bumps and the “substrate-side” grating resonance did not respond appreciably to refractive index changes, and could therefore be used as a reference.

As a measure of the performance as plasmonic sensors, it is necessary to measure both the bulk (i.e. water vs. another fluid) and the local (i.e. a thin biological film) refractive index sensing response. To measure the bulk sensing characteristics of our grating structures, a microfluidic channel was placed on top of the device and solutions with varying indices of refraction were injected sequentially. The spectral position of the peak at 700 nm was tracked as the index varied and is shown in Figure 6a. The bulk shift of the resonance is measured to be 410 nm per refractive index unit (RIU). For comparison, data for the spectral dip at 726 nm has also been plotted. In most sensing experiments it is also important to have local sensitivity within a few nanometers, since that is where any molecular interactions are occurring. Therefore, to characterize the local sensitivity, thin layers of Al_2O_3 were added in several discrete steps via atomic layer deposition (ALD) [57,58]. Steps starting at about 2.5 nm and increasing to 8 nm were taken up to 50 nm of Al_2O_3 . The spectral position of the peak at 700 nm was once again followed (Figure 6b) and the device shows a linear shift of 1.1 nm per nm of Al_2O_3 . These experiments are done in water, giving a slightly lower sensitivity than similar layer-by-layer measurements done in air. This is expected since the refractive index contrast between Al_2O_3 ($n = 1.65$) and air is greater than that between Al_2O_3 and water. As another method to characterize the local sensitivity, a single spectrum was measured both before and after the sensor was immersed in a 1 nM solution of dodecanethiol in ethanol to form a self-assembled monolayer on the surface, showing a spectral shift of 1.5 nm.

Many devices that use plasmon resonances for sensing depend on optical measurements that are taken through the liquid being studied. Such setups require that the sensing solutions of interest be optically clear. In contrast, our devices decouple the liquid and the optical sides, allowing for the use of opaque solutions. To demonstrate sensing with opaque liquids, various concentrations of food coloring dye (FD&C Red 40) were injected through the microfluidic channels, as shown in Figure 6c, and the change in the spectral peak position was monitored as shown in Figure 6d. It can be seen that as the concentration of food coloring was increased, there was an increase in the bulk refractive index and a red shift in the spectrum. There was not, however, significant degradation or weakening of the optical signal as the solution became more opaque. Additionally, to demonstrate real-time sensing capabilities using an opaque and highly scattering sample, a solution of milk was injected into the microfluidic channel. A single spectrum was taken every 2.7 seconds, and the position of the peak was continually monitored. Figure 6e shows the shift in the resonance with time. The large shift seen when the milk is injected is mostly due to the bulk index change of the milk. When water is injected to wash the surface, a net shift of about 0.5 nm is seen from adsorption of milk proteins on the Au surface. Such measurements using opaque solutions cannot be performed using transmission-mode nanoplasmonic sensors and demonstrates the advantage of our buried nanoplasmonic structures for biosensing.

In conclusion, we have demonstrated a unique sensing architecture using plasmonic gratings buried underneath an ultrasmooth and ultrathin Au sensing surface. These devices retain several advantages of conventional prism-based SPR by operating in a reflection-mode geometry. This decouples the optical and the fluidic access, allowing the use of opaque or highly scattering media. By using the embedded plasmonic nanostructures, optical tunability and normally incident epi-illumination and collection are also possible. Ultrasmooth surfaces will be particularly useful for studying the biophysics of lipid membranes, membrane-bound receptors, and lipid rafts. Furthermore, various other structures can be embedded that may have applications in on-chip nanophotonic circuitry [44], lithography, and plasmonic trapping [45].

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References

1. Brolo AG, Gordon R, Leatham B, Kavanagh KL. *Langmuir*. 2004; 20:4813–4815. [PubMed: 15984236]
2. Dahlin AB, Zäch M, Rindzevicius T, Käll M, Sutherland DS, Höök F. *J Am Chem Soc*. 2005; 127:5043–5048. [PubMed: 15810838]
3. Homola J. *Chem Rev*. 2008; 108:462–493. [PubMed: 18229953]
4. Anker JN, Hall WP, Lyandres O, Shah NC, Zhao J, Van Duyne RP. *Nature Mater*. 2008; 7:442–453. [PubMed: 18497851]
5. Novotny L. *Phys Today*. 2011; 64:47–52.
6. Atwater HA, Polman A. *Nature Mater*. 2010; 9:205–213. [PubMed: 20168344]
7. Xu H, Käll M. *Phys Rev Lett*. 2002; 89:246802. [PubMed: 12484969]
8. Haynes CL, McFarland A, Van Duyne RP. *Anal Chem*. 2005; 77:338A–346A.
9. Coe JV, Heer JM, Teeters-Kennedy S, Tian H, Rodriguez KR. *Annu Rev Phys Chem*. 2008; 59:179–202. [PubMed: 17988200]
10. Challener WA, Peng C, Itagi AV, Karns D, Peng W, Peng Y, Yang X, Zhu X, Gokemeijer NJ, Hsia YT, Ju G, Rottmayer RE, Seigler MA, Gage EC. *Nature Photon*. 2009; 3:220–224.
11. Ritchie R. *Phys Rev*. 1957; 106:874.
12. Homola J, Yee SS, Gauglitz G. *Sens Actuators, B*. 1999; 54:3–15.
13. Gordon R, Sinton D, Kavanagh KL, Brolo AG. *Acc Chem Res*. 2008; 41:1049–1057. [PubMed: 18605739]
14. Liedberg B, Nylander C, Lundstrom I. *Biosens Bioelectron*. 1995; 10:R1–R9.
15. Rindzevicius T, Alaverdyan Y, Dahlin AB, Höök F, Sutherland DS, Käll M. *Nano Lett*. 2005; 5:2335–2339. [PubMed: 16277479]
16. Tetz K, Pang L, Fainman Y. *Opt Lett*. 2006; 31:1528–1530. [PubMed: 16642161]
17. Lesuffleur A, Im H, Lindquist NC, Oh SH. *Appl Phys Lett*. 2007; 90:243110.
18. Yang JC, Ji J, Hogle JM, Larson DN. *Nano Lett*. 2008; 8:2718–2724. [PubMed: 18710296]
19. Eftekhari F, Escobedo C, Ferreira J, Duan X, Giroto EM, Brolo AG, Gordon R, Sinton D. *Anal Chem*. 2009; 81:4308–4311. [PubMed: 19408948]
20. Masson JF, Murray-Methot MP, Live LS. *Analyst*. 2010; 135:1483–1489. [PubMed: 20358096]
21. Yanik AA, Huang M, Kamohara O, Artar A, Geisbert TW, Connor JH, Altug H. *Nano Lett*. 2010; 10:4962–4969.
22. Nakamoto K, Kurita R, Niwa O. *Anal Chem*. 2012; 84:3187–3191. [PubMed: 22283116]
23. McFarland AD, Van Duyne RP. *Nano Lett*. 2003; 3:1057–1062.
24. Sonnefraud Y, Verellen N, Sobhani H, Vandenbosch GAE, Moshchalkov VV, Van Dorpe P, Nordlander P, Maier SA. *ACS Nano*. 2010; 4:1664–1670. [PubMed: 20155967]
25. Gao Y, Gan Q, Xin Z, Cheng X, Bartoli FJ. *ACS Nano*. 2011; 5:9836–9844. [PubMed: 22067195]
26. Feng J, Siu VS, Roelke A, Mehta V, Rhieu SY, Palmore GTR, Pacifici D. *Nano Lett*. 2012; 12:602–609. [PubMed: 22200183]
27. Smith E, Corn RM. *Appl Spectrosc*. 2003; 57:320A–332A.

28. Piliarik M, Vaisocherová H, Homola J. *Biosens Bioelectron.* 2005; 20:2104–2110. [PubMed: 15741081]
29. Shumaker-Parry JS, Aebbersold R, Campbell CT. *Anal Chem.* 2004; 76:2071–2082. [PubMed: 15053673]
30. Dostálek J, Homola J, Miler M. *Sens Actuators, B.* 2005; 107:154–161.
31. Nagpal P, Lindquist NC, Oh SH, Norris DJ. *Science.* 2009; 325:594–597. [PubMed: 19644116]
32. Spinke J, Yang J, Wolf H, Liley M, Ringsdorf H, Knoll W. *Biophys J.* 1992; 63:1667–1671. [PubMed: 19431869]
33. Cooper MA. *J Mol Recognit.* 2004; 17:286–315. [PubMed: 15227637]
34. Hegner M, Wagner P, Semenza G. *Surf Sci.* 1993; 291:39–46.
35. Naumann R, Schiller SM, Giess F, Grohe B, Hartman KB, Kärcher I, Köper I, Lübken J, Vasilev K, Knoll W. *Langmuir.* 2003; 19:5435–5443.
36. Rueda A, Vogel N, Kreiter M. *Surf Sci.* 2009; 603:491–497.
37. Lindquist NC, Nagpal P, Lesuffleur A, Norris DJ, Oh SH. *Nano Lett.* 2010; 10:1369–1373. [PubMed: 20235511]
38. Jose J, Segerink FB, Korterik JP, Gomez-Casado A, Huskens J, Herek JL, Offerhaus HL. *J Appl Phys.* 2011; 109:064906.
39. Im H, Lee SH, Wittenberg NJ, Johnson TW, Lindquist NC, Nagpal P, Norris DJ, Oh SH. *ACS Nano.* 2011; 5:6244–6253. [PubMed: 21770414]
40. Lindquist NC, Johnson TW, Norris DJ, Oh SH. *Nano Lett.* 2011; 11:3526–3530. [PubMed: 21834564]
41. Vogel N, Zieleniecki J, Köper I. *Nanoscale.* 2012; 4:3820–3832. [PubMed: 22628064]
42. Lindquist NC, Nagpal P, McPeak KM, Norris DJ, Oh SH. *Rep Prog Phys.* 2012; 75:036501. [PubMed: 22790420]
43. Love JC, Estroff LA, Kriebel JK, Nuzzo RG, Whitesides GM. *Chem Rev.* 2005; 105:1103–1170. [PubMed: 15826011]
44. Ebbesen TW, Genet C, Bozhevolnyi SI. *Phys Today.* 2008; 61:44–50.
45. Righini M, Zelenina AS, Girard C, Quidant R. *Nature Phys.* 2007; 3:477–480.
46. Castellana ET, Cremer PS. *Surf Sci Rep.* 2006; 61:429–444.
47. Lezec HJ, Degiron A, Devaux E, Linke R, Martín-Moreno L, García-Vidal FJ, Ebbesen TW. *Science.* 2002; 297:820. [PubMed: 12077423]
48. Ouellet E, Lausted C, Lin T, Yang CWT, Hood L, Lagally ET. *Lab Chip.* 2010; 10:581–588. [PubMed: 20162233]
49. Lee SH, Johnson TW, Lindquist NC, Im H, Norris DJ, Oh SH. *Adv Funct Mater.* 2012; 22:1002/advf.201200955
50. Teperik TV, Popov VV, Garcia de Abajo FJ, Abdelsalam M, Bartlett PN, Kelf TA, Sugawara Y, Baumberg JJ. *Opt Express.* 2006; 14:1965–1972. [PubMed: 19503527]
51. Barnes WL. *J Opt A: Pure Appl Opt.* 2006; 8:S87–S93.
52. Klar T, Perner M, Grosse S, von Plessen G, Spirkel W, Feldmann J. *Phys Rev Lett.* 1998; 80:4249–4252.
53. Giudicatti S, Marabelli F, Valsesia A, Pellacani P, Colpo P, Rossi F. *J Opt Soc Am B.* 2012; 29:1641–1647.
54. Burke J, Stegeman G, Tamir T. *Phys Rev B.* 1986; 33:5186–5201.
55. Kelf T, Sugawara Y, Cole R, Baumberg JJ, Abdelsalam M, Cintra S, Mahajan S, Russell A, Bartlett P. *Phys Rev B.* 2006; 74:245415.
56. Sannomiya T, Scholder O, Jefimovs K, Hafner C, Dahlin AB. *Small.* 2011; 7:1653–1663. [PubMed: 21520499]
57. Whitney AV, Elam JW, Zou S, Zinovev AV, Peter C, Schatz GC, Van Duyne RP. *J Phys Chem B.* 2005; 109:20522–20528. [PubMed: 16853656]
58. Im H, Lindquist NC, Lesuffleur A, Oh SH. *ACS Nano.* 2010; 4:947–954. [PubMed: 20131870]

Figure 1.

Various plasmonic sensing schemes. (a) Kretschmann prism coupling. The optical and fluidic paths are decoupled and the sensing surface is topographically flat, but with an oblique incidence angle. (b) Nanoparticles and localized SPR sensing. The sensing surface has topography and is often illuminated through the fluid with a dark-field objective. (c) Nanohole array SPR sensing. Surface plasmons are excited with normally-incident light for high-resolution imaging. The sample has topography and the light passes through the fluid. (d) The work presented here with backside SPR detection with an ultrasmooth sensing surface. The normally incident light is decoupled from the fluid in a reflection-mode setup.

Figure 2. Fabrication process. (a) A thin Au layer is first deposited on a flat Si wafer template. (b) A second, patterned, Au layer is then added. (c) Applying an adhesive backing layer and (d) stripping the device reveals the ultrasmooth surface with the grating now buried underneath. (e) The final ultrasmooth surface with buried gratings.

Figure 3.

(a) Cross-sectional scanning electron micrograph (SEM) of the completed device. (b) Atomic force microscopy (AFM) scans of the top-side (after template stripping) and the grating side of the device (before template stripping). The template-stripped Au surface shows less than 5 Å RMS surface roughness. (c) AFM scan of the top surface after template stripping and (d) the grating before the template stripping procedure. Horizontal lines show the cross-sections for panel (b).

Figure 4. Sample buried plasmonic devices. (a) A microscope image of an array of buried linear gratings. The light is reflected from the backside (i.e. through the epoxy) of the device. SEM of a linear grating structure (b) before and (c) after template stripping. (d) A buried bull's eye after template stripping and (e) a buried nanodot array after template stripping, both exemplifying the versatility of the fabrication process presented here.

Figure 5.

Characterization of the optical reflection spectra. (a) A single spectrum from a device with a period of 500 nm and a grating line width of 200 nm is shown to match (b) the FDTD simulations. The origin of the resonances, as discussed in the text, are studied in (c) which shows simulations of the z -component of the electric field distribution at each of the three reflection dips in the spectrum shown in (b). The arrows indicate the direction of normally incident x -polarized light with an illumination amplitude of unity. (d) As the period of the grating changes, resonance shifts are seen experimentally and (e) from the simulations. The black dashed lines indicate the locations of grating resonances in the thin gold film and the dashed ellipse indicates the localized resonance of the grating bumps. The reflection spectra from the buried gratings in (a, b) were normalized to a single reflection spectrum from an unpatterned 20 nm thick gold film. The colormaps in (d, e) were also normalized in this manner.

Figure 6.

Sensing with the grating device. (a) Bulk sensitivity measurements of the peak at 700 nm with varying concentrations of water/glycerol, showing a bulk sensitivity of 410 nm RIU⁻¹. Data for the dip at 726 nm is also shown. (b) Conformal layers of ALD were added to the sensor to test local sensitivity and the peak resonance shifted 1.1 nm per nm of Al₂O₃. (c) A photograph of a sample microfluidic chip. Due to our backside geometry, colored and/or opaque and highly scattering liquids can be used. (d) Spectra taken with varying concentrations of food coloring dye in water showing a shift due to the change in the bulk refractive index. (e) Real-time sensing of milk proteins non-specifically binding to the Au surface. These results demonstrate an ability for real-time sensing in opaque or highly-scattering liquids.