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Substrate effects on surface plasmons in single nanoholes

Kevin L. Shuford [a,*], Stephen K. Gray [b], Mark A. Ratner [c], George C. Schatz [c]

[a] Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6142, United States
[b] Chemistry Division, Argonne National Laboratory, Argonne, IL 60439, United States
[c] Department of Chemistry, Northwestern University, Evanston, IL 60208-3113, United States

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Abstract

Light transmission and electric field enhancement are calculated for a cylindrical aperture in a silver slab resting on a glass substrate. We find that these properties are influenced significantly by the presence of the substrate. The results suggest that variations in the local dielectric alter the properties of localized surface plasmons that drive the field enhancement and transmission.

Manipulating the flow of electromagnetic energy is of great interest for nanophotonic applications. Much of the current research in the field followed the finding that arrays of subwavelength apertures in metal films can transmit significantly more light through the holes than is expected from geometric optics [1]. The effect has been studied both experimentally and theoretically providing the generalized mechanism of plasmon mediated enhancement [1–4]. The light harvesting properties and intense local field enhancements also make nanoholes good systems for surface enhanced spectroscopies such as Raman [5,6] and fluorescence [7,8]. In some cases, these unique nanocontainers permit the measurement of single molecule properties [9,10].

In this Letter, we present light transmission and field enhancements relevant to surface enhanced Raman spectroscopy for single nanoholes in a silver slab. Particular emphasis is placed on the effects produced when adding a substrate. While one might imagine that the effect of the substrate involves simple dielectric shifts that are easily understood, we find that the transmission spectra are more complex, showing much sharper resonance structures that have no counterpart in the vacuum case. To understand this behavior, we study the relationship between the hole transmission results and the corresponding scattering for a particle that matches the size of the hole. This analysis is based on the (somewhat imprecise) use of Babinet's principle, in which the optical properties of particles and holes are expected to be similar, particularly with respect to localized plasmon resonance excitation. This analogy has been noted before [11], and in the present application enables us to understand the variation in resonance structure with substrate dielectric constant. We recognize that holes exhibit both localized surface plasmon (LSP) and propagating surface plasmon polarization (SPP) excitations, so the Babinet analogy is imperfect; however, we shall see that it is nevertheless still useful.

Our theoretical treatment for solving Maxwell's equations in cylindrical coordinates using the Finite-Difference Time-Domain method has been reported previously [12,13]. Here we treat the case where the incident field propagates toward the slab first contacting the vacuum/silver interface followed by the silver/substrate interface. The polarization of the incident field is chosen to be normal to the symmetry axis of the aperture.

Fig. 1 shows the calculated transmission for a 100 nm thick silver film for various hole sizes. Normalization in this figure is to the geometrical area of the hole so unit transmission corresponds to the geometrical optics limit. As expected, the amount of light transmitted increases as the hole gets larger. In vacuum, a sizeable red-shift occurs in the transmission as the size of the aperture is increased.

* Corresponding author.
E-mail address: shufordkl@ornl.gov (K.L. Shuford).

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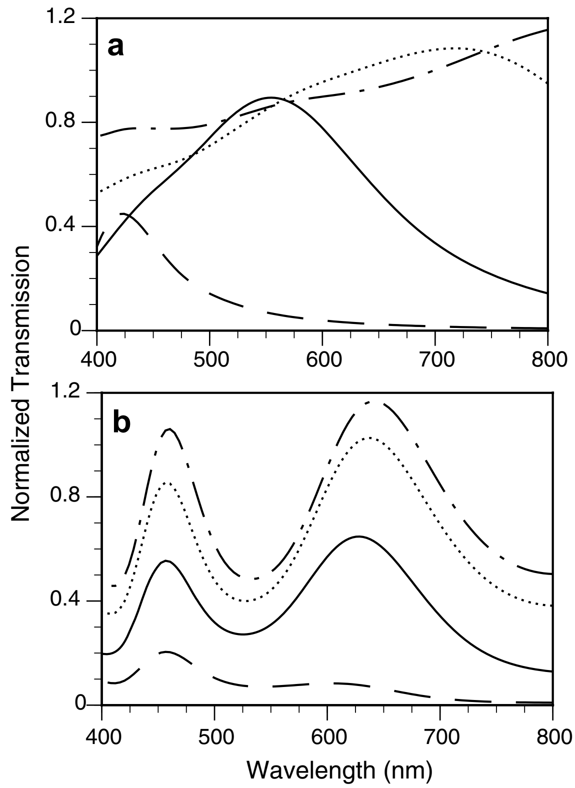

Fig. 1. Transmission through apertures in a 100 nm silver slab. The dashed, solid, dotted, and dot-dashed lines correspond to hole diameters of 100, 200, 300, and 400 nm, respectively. Panel a is in vacuum, and Panel b includes a glass substrate.

For the larger apertures a small shoulder is present around 450 nm in addition to the main transmission peak. Light transmission is altered significantly by the addition of a glass substrate (relative dielectric constant $\epsilon = 2.31$). Two, sharper peaks are present for each hole size around 450 and 625 nm. Both red-shift slightly as the hole diameter is increased, with the longer wavelength feature displaying a more pronounced shift.

To understand the trends observed in the transmission spectra, we consider the optical properties of disk-shaped nanoparticles under similar excitation conditions. Fig. 2 shows the scattering efficiency of silver disks calculated using the Discrete Dipole Approximation [14]. As the diameter of the disk increases, the peaks red-shift and broaden. Shoulders and local maxima are observed for the 200 and 300 nm particles in the 400–500 nm range of wavelengths. Examination of the induced polarization and electric field intensity of the nanoparticle indicates that these features correspond to quadrupole excitations, thus confirming the expected Babinet result for the case of no substrate. Also shown is the spectrum for a disk in glass. The peaks sharpen and shift to longer wavelengths compared to the vacuum results. Note that the quadrupole shift is approximately half of the dipole shift as the medium is changed from vacuum to glass, demonstrating that higher-order excitations are less sensitive to variations in

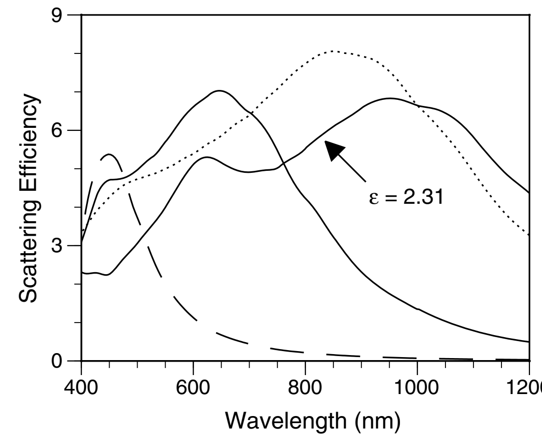

Fig. 2. Scattering of 100 nm thick silver disks. The dashed, solid, and dotted lines correspond to diameters of 100, 200, and 300 nm, respectively. The medium is vacuum except where indicated.

size and medium than dipole excitations. The transmission peaks in Fig. 1a tend to be blue-shifted from the corresponding nanoparticle resonances in Fig. 2, yet evolve in a similar manner as the diameter is increased.

We now examine the variation of the transmission spectra with substrate permittivity to understand the plasmon excitation when the substate is present in Fig. 1b. Fig. 3 displays the transmission for a 300 nm hole for dielectric constant values ranging from vacuum to glass. The maximum transmission in vacuum (which we can assign to excitation of the dipole LSP) occurs at 720 nm, and shifts beyond 800 nm as the dielectric constant is increased. Features below 400 nm and the shoulder at 450 nm in vacuum sharpen and red-shift to become the prominent transmission peaks when $\epsilon = 2.31$. The splitting between these features varies very little as the permittivity increases even though the substrate alters the local dielectric environment at one interface resulting in frequency shifts. The lack of asymmetry in the peak evolution indicates that we are sampling plasmonic states residing on each side of the film that

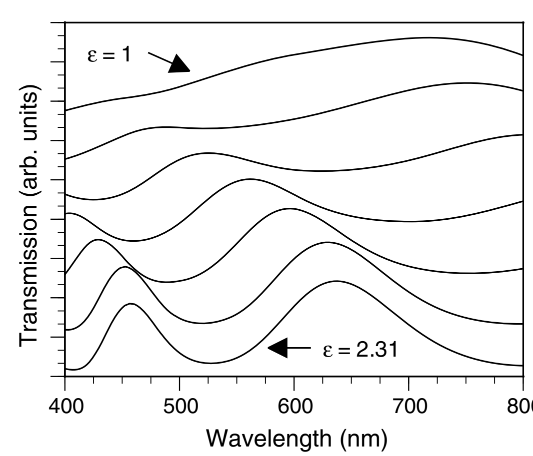

Fig. 3. Transmission through a 300 nm aperture in a 100 nm slab. The dielectric constant of the substrate is increased from 1 to 2.25 in 0.25 increments, and the $\epsilon = 2.31$ result is also included.

are strongly coupled to one another. The transmission properties are similar to nanoparticle scattering (Fig. 2) upon varying the medium from vacuum to glass. This suggests that the global maxima in Fig. 1a correspond to localized dipole excitations at the rim, and the peaks in Fig. 1b originate primarily from higher-order excitations ($n = 2$ and $n = 3$ multipoles) that have been red-shifted by the presence of the substrate. The fact that the transmission peaks are not shifted greatly upon increasing the hole diameter further supports the assignment of higher-order excitations in accord with the nanoparticle analogy. We should note that nanohole arrays have been used in chemical sensing applications [15]. Fig. 3 shows that even isolated hole structures produce detectable shifts in peak positions in response to small changes in the dielectric environment.

Previous calculations in vacuum have shown that the maximum electric field enhancement is localized at the aperture rim along the direction of the incident polarization [13]. These studies also found that the wavelength of maximum enhancement at the hole entrance was slightly red-shifted from that of the hole exit, and that the transmission maximum (see Fig. 1a) was typically between these values. Fig. 4 shows that the glass substrate significantly alters the properties of the nanohole fields. Enhancement at the hole entrance is still red-shifted from the exit, but the shift is considerably less than in vacuum. The transmission peaks now correlate well with the field enhancements on the exit side, whereas in vacuum the transmission peaks were slightly blue-shifted. It can be seen that 100 nm aperture generates sizeable local field enhancements on the entrance side of the slab, but has a much smaller effect on the exit side. This is consistent with the small transmission for that hole size, and it indicates that a LSP is not excited significantly on the exit side of the film. Larger apertures allow the incident field to penetrate more easily and induce coupling, which produces larger field enhancements on the exit side and leads to greater transmission of light. We find similar results for 300 nm thick slabs for apertures greater than 200 nm. Holes smaller than this do not allow enough coupling between the entrance and exit locations due to the thicker slab, and yield small transmission values and enhancements on the exit side. The magnitude of the field enhancements at the hole rim is roughly a factor of 100. This is similar to what we found previously for holes in vacuum [13], and is consistent with estimates from recent SERS measurements [6]. This indicates that although the plasmon resonances are sharper for the film on a glass substrate than in vacuum, the local field enhancement is comparable.

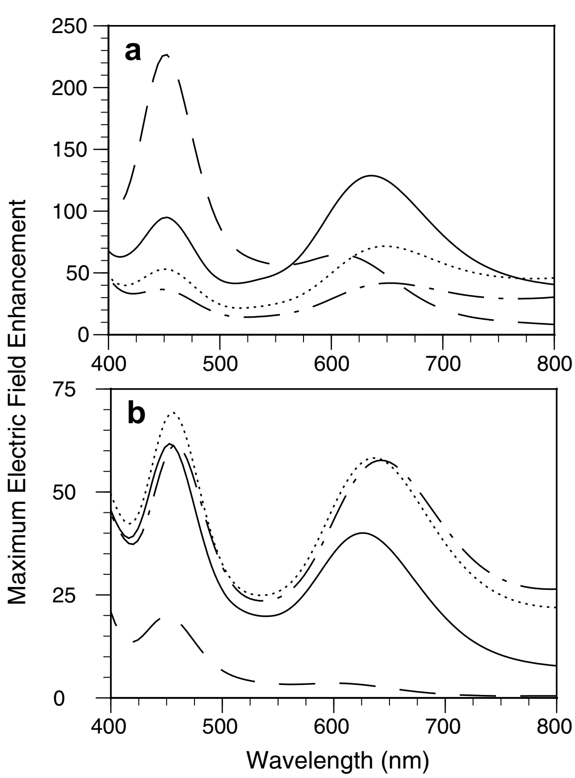

Fig. 4. Maximum field enhancement in a 100 nm silver slab on a glass substrate. The dashed, solid, dotted, and dot-dashed lines correspond to hole diameters of 100, 200, 300, and 400 nm, respectively. Panel a is at the hole entrance, and Panel b is at the hole exit.

In conclusion, we have shown that the light transmission and field enhancement in nanohole systems are influenced greatly by a substrate. Factors such as aperture size, slab thickness, and local dielectric environment significantly alter these properties. In the absence of arrays or gratings, the transmission characteristics and field enhancements are dominated by LSP excitations of the hole. We anticipate these results being useful for future nanophotonic devices, chemical sensing applications, and surface enhanced spectroscopies.

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