Optical resonances in periodic surface arrays of metallic patches


The transmission of light along the surface normal through an air–quartz-glass interface covered with a periodic array of thin, rectangular gold patches has been studied over the visible to infrared range. The various structures that are observed can be qualitatively understood as arising from standing-wave resonances set by the size and surroundings of the metal patches. A method-of-moments calculational scheme provides simulations in good quantitative agreement with the data. It is shown how the standing-wave picture provides a useful conceptual framework to understand and exploit such systems.

1. Introduction

In this paper the interaction of light with a layer of conducting particles is studied. The specific, simple system under consideration is illustrated in Fig. 1 where the particles are thin, rectangular patches of metal, fixed in a periodic rectangular array on a transparent, flat substrate. Our interest is in understanding the possible optical resonances that can occur when light (polarized along the long patch dimension, for example) is incident along the surface normal. Over narrow ranges of (vacuum) wavelength \( \lambda \), there will be enhanced reflection and absorption, with a corresponding reduction of transmission.

The interpretation of the physics involved in such resonances depends on the relative size of \( \lambda \) and the long dimension \( w_y \). We focus here on the spectral range where \( \lambda \) is comparable to \( w_y \). As a first approximation, consider a single patch in isolation. It can be viewed as an antenna that supports various standing waves. The simplest estimate for the location of these is \( j(\lambda/2) = w_y \), where \( j \) is an integer. The strongest resonance is the first, \( j = 1 \), or dipole resonance. Its associated current profile has nodes at each end of a patch and a single antinode in the center. This sort of picture is most reasonable in the infrared and longer-wavelength range, where metals behave as (near-) perfect conductors. Many systems have been studied in this limit, usually under the name of frequency-selective surfaces.\(^1\)\(^-\)\(^3\) More recent research has been moving toward shorter wavelengths.\(^4\)\(^-\)\(^5\)

Of course the resonance locations also depend on the other parameters of the model such as the remaining dimensions of the patch, width \( w_z \), and thickness \( h \); the periods of the surface array \( d_y \) and \( d_z \); the optical index of the glass substrate \( n_g = [\epsilon_g/\epsilon_0]^{1/2} \); and the dielectric response of the metal. Several studies have compared experiment and theory with regard to these dependencies,\(^6\)\(^-\)\(^11\) but they have concentrated mostly on the dipole resonance. Here we examine a series of resonances to learn how well they can be understood in the standing-wave picture. As the mode index \( j \) is increased (or \( w_y \) is decreased), the resonance wavelength will decrease and the material dependence of the metal’s response should become more important. Indeed, with \( w_y \sim 1 \mu m \), one readily encounters resonance frequencies comparable to the bulk plasma frequency as \( j \) increases.
To study a series of resonances one needs to measure over a broad spectral range. This is accomplished by use of different instruments for the visible and infrared ranges. In Section 2 we describe these measurements along with our sample preparation methods. The theoretical model for calculations of the system’s optical response is outlined in Section 3 where we generalize an approach developed earlier to simulate spectra near dipole resonances in the infrared.\textsuperscript{10} The theory predicts a whole transmission spectrum, $T$ versus $\lambda$, and hence can be directly compared with experimental data. Finally, in Section 4 we summarize what has been learned by such comparisons and note several implied features that deserve further study. We conclude that a standing-wave picture does provide useful insights, and our detailed simulations do agree well with the measurements.

2. Sample Fabrication and Measurement
The basic sample configuration is shown in Fig. 1. The metal patches are made of gold, deposited by electron-beam lithography in Graz, Austria, on a quartz-glass substrate 1 mm thick that was doped with indium tin oxide over a depth of 3 nm. Details on this technique can be found in Ref. 12. For the measurements described here only the long dimension of the metal patches was varied. The other parameters were fixed at the nominal values of $w_z = 91$ nm, $h = 17$ nm, $d_y = 2.11$ $\mu$m, and $d_z = 0.54$ $\mu$m. These geometric parameters have an uncertainty of several percent.

Because the total extent for a particular pattern is a 150-\(\mu\)m square, special care is necessary to measure the optical response. In the visible range, extinction spectra of linearly polarized light are measured with a Carl Zeiss MMS1 spectrometer coupled to a conventional optical microscope equipped with a low-numerical-aperture (0.075) objective.

Hence the greatest deviation from normal incidence of any light that can be collected is 4.3°.

Results for a set of patterns of varying $w_y$ are shown in Fig. 2. The polarization of the incident light is along the $y$ direction and the $T_0$ that appears in the extinction definition is the transmission through a region of the substrate where there are no metal patches. Working with $T/T_0$ provides a convenient normalization that suppresses variations independent of the patch pattern, which are not of interest here. The resonances of interest are associated with the extinction peaks. These are evident only for $\lambda \approx 700$ nm; in earlier studies with Ag patches, peaks were observed down to 500 nm.\textsuperscript{13}

Measurements in the infrared of the same samples were made with the Perkin-Elmer Fourier-Transform Spectrum 2100 at Agere Systems in Orlando, Florida. The incident light was unpolarized, and because it was focused with an $F/3.2$ lens into a 100-\(\mu\)m-diameter spot through the center of a pattern on the surface, its path could deviate by up to 9° away from the surface normal. Again we work with only the relative transmission $T/T_0$. Plots of $T/T_0$ versus $\lambda$ for different choices of $w_y$ are given in Section 3 where they are compared with the simulations.

3. Theoretical Model
The calculational scheme we use is based on a specialization of the general formalism that has been developed for frequency-selective surfaces.\textsuperscript{1} The main ideas have been described earlier,\textsuperscript{10,11} so we discuss only modifications that were made for the present study. We concentrate on the transmission coefficient for a beam to cross the patterned vacuum–glass interface without deflection. Multiple reflections within the glass are ignored. It was demonstrated in Ref. 11 that such processes are of little quantitative significance. We also ignore any
possible optical effects of the indium tin oxide doping layer. The metal patches are treated as negligibly thin but possessing a complex-valued sheet resistance $R = 1/\Sigma$. The conductance $\Sigma$ is calculated from

$$Z_0\Sigma = -2\pi i(\epsilon/\epsilon_0 - 1)\frac{h}{\lambda},$$

(1)

where $\epsilon$ is the dielectric function of the metal and $Z_0 = [\mu_0/\epsilon_0]^{1/2} \approx 377$ $\Omega$ is the impedance of free space. If the metal's response at frequency $\omega$ were described simply by a conductivity $\sigma$, then $\epsilon = \epsilon_0 + i\sigma/\omega$ and $\Sigma$ would be $\sigma h$. For gold we start with the Drude form

$$\epsilon = \epsilon_d - \epsilon_0\omega_p^2/\left[(\omega + i/\tau)\right],$$

(2)

where $\epsilon_d/\epsilon_0 = 8$ characterizes the polarization of the d electrons and the free-electron response is described by the plasma frequency $\hbar\omega_p = 8$ eV and a scattering rate $1/\tau$. Such a functional form provides a reasonable fit to the bulk optical data of gold, if we use $1/\omega_p\tau = 0.008$, and gives us the freedom to tweak $\epsilon$ with physically understandable parameters. In particular we show below that $1/\tau$ for the thin metal patches must be significantly increased if our theory is to reproduce the transmission data for the patterned surfaces.

In earlier calculations with the incident electric polarization along the $y$ direction, we constrained the induced current in the metal patches to be only along $\hat{y}$ and to be independent of $z$. Here we allow for a more general (but still two-dimensional) current pattern. Again, with the incident electric field along $\hat{y}$, we write for the current density within each patch

$$J_y(y, z) = \sum_{j,k} d_{j,k}^{(y)} \cos\left(j\frac{\pi}{2}\hat{y}\right) \cos\left(k\frac{\pi}{2}\hat{z}\right),$$

(3)

$$J_z(y, z) = \sum_{j,k} d_{j,k}^{(z)} \sin\left(j\frac{\pi}{2}\hat{y}\right) \sin\left(k\frac{\pi}{2}\hat{z}\right),$$

(4)

where $\hat{y} = 2y/w_y$ and $\hat{z} = 2z/w_z$ with the origin for $y$ and $z$ in the center of a patch. The $j$'s and $k$'s are nonnegative integers with the $j$'s odd and the $k$'s even. The value $k = 0$ is to be included in Eq. (3), but not in Eq. (4). With these choices, $J_y$ vanishes at the extreme values $|\hat{y}| = 1$, and $J_z$ vanishes at the extreme values $|\hat{z}| = 1$. On the other hand the current flow parallel to an edge ($J_y$ along $|\hat{z}| = 1$ or $J_z$ along $|\hat{y}| = 1$) is nonzero and can be enhanced. The symmetries of the currents under $y \rightarrow -y$ or $z \rightarrow -z$ are correct if the light is incident along the surface normal; otherwise basis functions of different parity need to be included. The expansion coefficients $d_{j,k}^{(y)}$ in Eqs. (3) and (4) are determined by the Galerkin version of the method of moments. With them the currents are known, and one can readily find the transmission coefficient of the beam moving along the surface normal into the glass substrate. The same relative transmission is obtained if the beam is incident from the glass, which in fact is the direction used in the measurements.

Typical results are shown in Fig. 3 for incident light polarized along $\hat{y}$. We use here (and below) a spline interpolation through tabulated values of $\epsilon_p/\epsilon_0$ for the quartz-glass substrate. The long dimension of the patch is $w_y = 1.10$ $\mu$m, and we plot the extinction as in Fig. 2 (but without a shift of 0.02). For the initial calculation we used the stated values of all the parameters defined above. However, the resulting peaks are too strong and sharp. The form of the peaks is better reproduced if we increase the Drude scattering rate by approximately a factor of 3. To better match the peak locations, we reduced the patch thickness from its nominal value $h = 17$ nm to $h = 16$ nm. The direction of these two modifications is not unreasonable. One expects there to be extra (surface) scattering in the thin patches, and their vertical profile is somewhat rounded rather than rectangular. However, the specific values of $1/\tau$ and $h$ that we use were chosen simply to produce a better match with the data of Fig. 2. Because the Drude representation of $\epsilon$ in Eq. (2) becomes increasingly poor as one crosses the interband threshold, we stop our calculation at $\lambda = 500$ nm. However, there is a clear upturn in extinction for shorter wavelengths in Fig. 2, so we also calculated what our model would predict if the optical data of Johnson and Christy are used for $\epsilon$. It appears that the onset of additional absorption in bulk gold below 500 nm does account for the observed enhanced extinction.

Next we hold fixed all parameters except $w_y$ and calculate for Fig. 4 the analog of the measured Fig. 2. There is reasonable agreement between the two plots, particularly for the location and shape of the several peaks evident at longer wavelengths. For each of the calculated curves, we used $1/\omega_p\tau = 0.024$ and $h = 16$ nm plus the nominal values of the other parameters. The slight disagreement between the-
The response to light polarized along $\hat{z}$ is negligible through most of the infrared. A standing wave is still possible but its location is set by $w_y$, which is much smaller than most $w_y$ values and hence first appears in the visible at $\lambda = 629$ nm. The structure of Fig. 5 shows that measurements with unpolarized light in the infrared can be directly interpreted in terms of resonances with the driving field along $\hat{y}$. We use

$$T^{\text{un}} / T_0^{\text{un}} = \frac{1}{2} \left[ T^{y} / T_0^{y} + T^{z} / T_0^{z} \right]$$

$$= \frac{1}{2} \left[ T^{y} / T_0^{y} + 1 \right],$$

where the $y$ or $z$ superscripts describe directions of the incident polarization and un denotes unpolarized.

Now compare the experiment and theory in the infrared. Results for different patterns are shown in the different panels of Fig. 6. We stress that the parameter choices in the calculations are the same as for Fig. 4. Overall the agreement continues to be good, in particular for the location and depth of the main resonance. The theory also reproduces some of the smaller structures, e.g., in (a), (b), (c), and (d) at
\( \lambda = 1.32, 1.74, 1.52, \text{ and } 1.20 \ \mu \text{m} \), respectively. As for disagreements, we note three general features. First, the theoretical transmission for wavelengths longer than the main resonance location is less than the measured transmission. Second, most of the kinks that are due to threshold anomalies in the simulations do not appear in the measurements. Third, there are additional local minima in the data around 1.55, 1.35, 1.1, and 1.7 \( \mu \text{m} \), respectively, that are missing in the calculations.

The resolution of the last two kinds of puzzle might lie with the spread of incident angles in the measurements. As noted in Section 2, the infrared arrangement allows light at up to 9° away from normal incidence to be collected. For a rough estimate of the consequences of this spread on the simulations, we performed some calculations with an incident beam away from the normal, but confined to the \( x-y \) plane. Results are shown in Fig. 7. Note first that the threshold anomalies split and shift as the angle of incidence \( \theta \) is varied. This occurs because the criterion for their appearance in our special geometry is

\[
\frac{\epsilon}{\epsilon_0} \frac{2\pi}{\lambda} = \left| G + \frac{2\pi}{\lambda} \sin \theta \right| ,
\]

where \( \lambda \) is the vacuum wavelength, \( \epsilon \) is either \( \epsilon_0 \) or \( \epsilon_g \), and

\[
G = 2\pi \left( \frac{n_y}{d_y} \hat{y} + \frac{n_z}{d_z} \hat{z} \right)
\]

is a two-dimensional reciprocal lattice vector of the patch array with \( n_y \) and \( n_z \) integers. The net influence of threshold anomalies will be reduced by an average over incident angles. Note too that the minimum in \( T/T_0 \) near 2 \( \mu \text{m} \) shifts to longer wavelengths (and better agreement with the data) as \( \theta \) increases. This happens because of an accidental overlap of the \( \theta = 0 \) threshold anomaly at 2.11 \( \mu \text{m} \) with an isolated patch resonance. As \( \theta \) increases, the overlap and consequent distortion are reduced.
A further development with increasing $\theta$ is the appearance of additional resonance minima in between those present at $\theta = 0$. In Fig. 7, these extra minima occur at 1.17, 1.67, and perhaps 3.3 $\mu$m. They arise from the broken symmetry for $\theta \neq 0$ that leads to additional basis terms in the expansion of $J_y$ in Eq. (3) of the form $d_{j,k}^{(y)} \sin j(\pi/2)\gamma \cos k(\pi/2)\zeta$ where both $j$ and $k$ are even integers. Compared with the experiment, the new modes are appearing in approximately the right places, but it is not fair to draw a definite conclusion because the calculations have not averaged over all possible incident directions and it is difficult to differentiate the new minima from threshold anomalies and measurement noise.

4. Discussion

In our comparisons so far of experiment and theory we have made little reference to the standing-wave picture described in Section 1. The reason for this is because the utility of that picture lies primarily in the conceptual overview it provides rather than in its specific numerical estimates. More sophisticated models, such as the one described in Section 3, are needed to produce reliable quantitative results. Nevertheless it is helpful to have the picture in mind as one attempts to understand and predict possible behaviors.

The standing-wave index $j$ corresponds to the $j$ index appearing in Eq. (3). With incident light polarized along $\gamma$, the $d_{j,k}^{(y)}$ on resonance will be enhanced for a particular $j$. This is illustrated in Fig. 8 that plots the magnitude of the complex amplitudes $|d_{j,k}^{(y)}|$ versus $\lambda$ for several (odd) values of $j$. These were chosen because the transmission changes are controlled by the spatial average of the induced current. From Eq. (3) this involves

$$\int_{-1}^{1} dy \int_{-1}^{1} dz J_y(y, z) = \frac{8}{\pi} \sum_{j} d_{j,k}^{(y)}(-1)^{j-1}/j, \quad (8)$$

where the sum is over odd values of $j$.

Fig. 8. Expansion coefficients versus wavelength for $w_{y} = 1.10$ $\mu$m. The location of structures correlates well with the minima and threshold anomalies in the $T/T_0$ of Fig. 5.

Fig. 9. Possible surface dispersion of resonance locations $\epsilon = \hbar \omega$ versus wave vector $Q$. The points are extracted from the results of Figs. 2 and 6 by Eq. (9). Some even $j$ modes, excited off normal incidence in the infrared, are included. The long- (short-) dashed curves are the light lines for glass (vacuum). At frequencies above them light can propagate away from the surface. The solid curve is the (lowest) retarded surface plasmon for a homogeneous Au layer of thickness $h = 16$ nm between vacuum and glass.

![Image](image.png)

$\hbar \omega = \omega_{p}^{2}Qh\left(\frac{E_{0}}{E_{g} + E_{0}}\right)$, \quad (10)

and use our results to determine the dispersion of the effective wave established near the patch. Thus we are not trying (yet) to predict the effective wavelength, but instead we are checking whether there is a common dispersion curve $\omega(Q)$ for different $j$ and $w_{y}$ that underlies all the observed resonances. Results from the measurements of Figs. 2 and 6 are collected in Fig. 9, which demonstrates that Eq. (9) does allow the resonance locations of various orders to be scaled onto a single curve.

The appearance of Fig. 9 is appealing and raises the further question as to whether the effective dispersion relation can be derived. As a first guess consider the nonretarded surface plasmon produced by a thin homogeneous layer of free electrons located at the interface between two dielectrics. For wave vector $Q$ within the plane,$^{21}$

$$\omega^{2} = \omega_{p}^{2}Qh\left(\frac{E_{0}}{E_{g} + E_{0}}\right), \quad (10)$$

where

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where \( \omega_p \) is the plasma frequency of the free electrons in bulk, \( h \) is the thickness of the metal layer, and we assumed that the two bounding dielectrics are vacuum and glass. Equation (10) is qualitatively correct but quantitatively unreliable. We need to allow for at least for the dielectric response of gold [by using the \( \varepsilon \) of Eq. (2)] and to include retardation. An analytic result is then not possible but a numerical dispersion can be obtained by the scheme of Ref. 22. For wave vector \( Q \) within the plane, fields away from the interface vary as \( \exp[i(Qy - \omega t)]\exp(-Q|x|) \) where \( t \) is time, \( x \) is along the surface normal, and

\[
Q = \left( Q^2 - \frac{\omega^2}{c^2} \frac{\varepsilon}{\varepsilon_0} \right)^{1/2}, \tag{11}
\]

with \( \varepsilon = \varepsilon_0(\varepsilon_0) \) in glass (vacuum). If one perturbs the surface with such an incident wave holding \( Q \) fixed and varying \( \omega \), then the reflected wave will show a resonant peak as \( \omega \) crosses the surface mode.

Results from such a procedure were also plotted in Fig. 9 with a constant \( n_x = 1.43 \). For larger \( Q \) the calculated curve is significantly above the data points. Use of Eq. (10) makes the discrepancy worse. In the opposite limit, theory and experiment seem to merge, or perhaps cross, as \( Q \to 0 \). Because we are looking in the calculation for a mode bound to the surface, the theoretical curve cannot cross the light line of glass that is defined by the vanishing of \( Q \) in Eq. (11). However, the measured points are not so constrained and may indeed come from resonances in the continuum. The problem is that the patterned surface couples different wave vectors together so estimates of \( \omega(Q) \) based on models of a homogeneous metal layer supporting a single \( Q \) cannot be expected to succeed.23 Another way of appreciating the problem is to recall that it has been shown\textsuperscript{10,19} that the shapes and locations of resonances depend on the lattice constants of the patch array. This can be viewed as arising from threshold anomalies or more simply from the interactions of different patches by their electrodynamic fields.

The consequent distortions are larger for modes with smaller \( j \) because the standing-wave pattern influences the range of the near fields. Given all these points of view it is clear that one must expect some imprecision in the standing-wave picture.

There are other ways in which the utility and the limitations of the standing-wave picture appear together. Consider, for example, the response to light polarized along the short dimension of the patches. A first estimate would be to use \( Q_{10} \) with \( \omega_y = \omega_x \), so one would expect for our samples that the resonances would be independent of \( \omega_y \) and off the scale of Fig. 9 because the minimum \( Q \) would be \( \pi/(0.091 \mu m) = 34.5 (\mu m)^{-1} \). In Fig. 10 we compare experiment and theory for the extinction of \( \hat{z} \) incident polarization. We identify the single peak in all cases as arising from the \( j = 1 \) dipole excitation. The location of this peak is a weak monotonic function of \( \omega_y \) in the simulations but appears slightly nonmonotonic in the data. The relative heights of the peaks compare well, decreasing with the amount of metal on the surface. There is no evidence of higher-order peaks, probably because these would be at or beyond the threshold for \( d \)-band excitations.

The extinction data for different incident polarizations could be used to make an interesting check of the consistency of the standing-wave picture. Imagine producing the same \( Q \) in two different ways:

\[
j_y \frac{\pi}{w_y} = Q = j_z \frac{\pi}{w_z}. \tag{12}\]

Then one should expect that simply by switching between two orthogonal incident polarizations it would be possible to excite one or the other of two degenerate modes. In the standing-wave picture this novel situation arises whenever \( \omega_y/w_y \) is the ratio of small odd integers. We almost have such a case in Figs. 2 and 10 when the extinction peak for \( \omega_y = 0.32 \mu m \) and \( j_y = 3 \) nearly overlaps that for \( j_z = 1 \). A more focused study of this effect is postponed to a future study. We expect with the quantitative imprecision.
of the standing-wave picture that one will need a series of samples with \( \omega / \omega_c \) close to the required ratio from Eq. (12) to find a sample where the resonances for orthogonal polarizations exactly coincide.

To summarize, we have measured optical transmission over a wide spectral range through surfaces covered with a periodic array of metal patches. Detailed simulations can successfully reproduce the principal features of the data. In addition we have shown how a standing-wave picture of the resonances provides a helpful overview of the physics, aiding the interpretation and suggesting new ways to examine and exploit these systems.

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References
23. Of course one may include the coupling among surface wave vectors and, as shown in Section 3, good agreement is then found for the mode locations.