Alternative Plasmonic Materials and Chalcogenides for Photonic Applications

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Key References:

Outline

- Basics of photonic materials
- Plasmonics: basics
- Classical vs alternative materials
- Endfire coupling into plasmons
- Chalcogenides for Photonics
- Maximal Absorption by a Bare, Structured Thin Film
- Maximal Absorption by a Thin Film plus Reflector
- Theory
- Experiment
I concentrate on the task of optimizing transmission, reflection and absorption for optical structures.

Other important tasks exist in photonics—e.g., enhancing local field strengths for sensing, second and third harmonic generation, supercontinuum generation, wave interaction optimisation—e.g., SBS.

To do real designs one needs to start with optical constants as a function of wavelength.

For pure elements and some important compounds, the collection of volumes by Edmund Palik is a good print reference. The web reference http://luxpop.com/RefractiveIndexList.html gives important material data in digital form.

For non-stoichiometric materials and less used compounds, original references are needed.
LYCURGUS CUP, a Roman goblet dating from the fourth century A.D., changes color because of the plasmonic excitation of metallic particles within the glass matrix. When a light source is placed inside the normally greenish goblet, it looks red.
The best choice of material can rarely deliver the required optical properties.

Thus different materials must be combined together in an optimized structure.

The structure may be randomized or ordered, isotropic or anisotropic.

Much of my work has dealt with periodic structures, optimizing them to deliver desired optical characteristics, sometimes over a narrow bandwidth or range of angles, at other times over a wide range of angles.

Periodic structures can deliver surprisingly strong results, based on effects such as Wood anomalies, resonance anomalies, guided wave coupling and cut-off effects.
The book *Plasmonics: From Basics to Advanced Topics* contains an excellent summary of the field.

Surface plasmons are surface electromagnetic waves, existing generally on materials with negative permittivity.

Despite the name, they can be regarded in most important aspects in the framework of Maxwell’s equations, rather than Schrödinger’s equation.

They can be divided into two classes; propagating surface plasmons or Surface Plasmon Polaritons (SPP’s), which propagate along an extended metallic surface, and Localized Surface Plasmons (LSP’s), which exist on the surface of individual metal particles, or ensembles of such particles.

LSP’s have the longer history, being involved in the properties of stained or colored glass, while SPP’s have been systematically studied since the discover of Wood Anomalies in 1902.
Figure 7.6: (Left) An evacuated tubular solar collector module. (From [32].) (Right) The calculated (C) and experimental (Exp) reflectance curves of a Cu-SiO$_2$ cermet. (From [35].)
Wood Anomalies and Grating Absorption

- Wood Anomalies of metallic diffraction gratings were discovered by Robert W. Wood in 1902, and Lord Rayleigh in 1907 pointed out their connection with diffracted orders being diffracted parallel to the grating surface (“passing off”).

- They attracted much interest from experimentalists (Strong, C.H., Palmer) and theoreticians (von Ignatowsky, U. Fano, Hessel and Oliner) alike from their discovery until the late 60’s.

- By then, integral equation and differential methods for modelling grating diffraction on mainframe computers were becoming available.

- The most stringent test available for these was how well they could model Wood Anomalies.
Fig. 2.3 Efficiency curves for the order \(-1\) and total energy curves for three holographic gratings experimental measurements with period 1.210 nm and height 190 nm covered with three different metals-illuminated by p-polarized laser beams. a Experimental measurements by Hutley and Bird; b results obtained from the new integral theory. a is reprinted by permission from Taylor & Francis Ltd (http://www.informaworld.com); [24] pp. 772–776
2. Theory of Wood's Anomalies

Fig. 2.20  Theoretical (a) and experimental (b) reflectance of a sinusoidal gold grating versus the angle of incidence for various groove depths $h$. Reprinted from [50], with permission from Elsevier.
Fig. 2.20 Theoretical (a) and experimental (b) reflectance of a sinusoidal gold grating versus the angle of incidence for various groove depths $h$. Reprinted from [50], with permission from Elsevier.
Fig. 1. (a) Spectral dependence of the 0-th order reflectivity of a shallow sinusoidal grating in normal incidence. Period $d = 0.62 \mu m$ and groove depth $h = 0.032 \mu m$, polarization TE, parallel (black) or TM, perpendicular (blue) to the groove direction. (b) Same as in (a) but for a 2D crossed grating representing the sum of two mutually perpendicular gratings identical to the one in (a). $d_x = d_z = 0.62 \mu m$, $h_x = h_z = 0.032 \mu m$, normal incidence. Order 0 in reflection - red, sum of orders 0 and -1 - in green, TM efficiency from Fig. 1a - blue squares.
Fig. 2. Comparison between the theoretical and experimental reflectivity in normal incidence of a grating similar to Fig.1, but having period $d_x = d_z = 0.6 \mu m$ and total groove depth $h = 0.08 \mu m$ ($h_x = h_z = 0.04 \mu m$). Theoretical results - black curve, experimental data - red squares.
The noble metals, chiefly gold and silver, are most used in plasmonics. In sensing for example, gold is well adapted to tethering of captor molecules, which link preferentially to other target molecules for sensing applications. Such metals are also well adapted to applications in which they function in reflection, with little field penetration to cause loss by absorption. However, in applications associated with transmission, or with long required propagation distances, absorption losses can be prohibitive. Attempts to compensate metallic loss by incorporating gain into the system have proven too difficult.
Metals are "too metallic": metal nitrides may be better for selected applications
Fig. 3. Real and imaginary parts of the relative permittivities of the four plasmonic materials: silver (Ag) [35], gold (Au) [35], titanium nitride (TiN) [24], and zirconium nitride (ZrN) [35].
In applications like sensing and high-density communication networks, it is necessary to couple efficiently a laser beam into surface plasmons.

A suitable method is end-fire coupling, in which the laser beam is aimed at the side of a dielectric-metal-dielectric waveguide structure.

This was studied theoretically in an early paper by Stegeman, Wallis and Maradudin, Opt.Lett., 8, 386-388 (1983).

We have studied end-fire coupling in detail in two recent papers: JOSA B, 32, 412-425 (2015) and 33, 1044-1054 (2016).
The theoretical model used consisted of a Gaussian beam approaching a metal-dielectric layered structure.

The domain was bounded by perfectly conducting walls.

Fields in the waveguide region were represented by a superposition of modes of the Kronig-Penney type.

The physical dimensions were such as to require large-scale modal superpositions (order 2000 modes).

As a result, a sophisticated mode finding method based on the Cauchy Residue Theorem was employed.

Accuracy of results was assessed using completeness and energy conservation conditions.
End-fire Coupling into Surface Plasmons-3

**Fig. 4.** SP coupling efficiencies $\eta$ of each plasmonic material when coupled into by an incident Gaussian beam centred about $x = \Delta l$ and with width $w = 2\lambda$.

**Fig. 5.** Maximum SP coupling efficiencies for optimised incident beam width and position, with $L_{\text{min}} = 0.5\lambda$. 
The results of the study were that end-fire coupling is a highly efficient technique, with coupling efficiencies of over 80% deliverable over a wide range of wavelengths.

The high efficiencies did not depend sensitively on incident beam positions, widths and centre position in relation to the structure.

Many results of this complicated theoretical method could in fact be explained using a simple formula based on maximising the overlap between the Gaussian beam and the target plasmon beam.

The alternative nitride materials could deliver higher coupling efficiencies than the noble metals silver and gold.

However, the propagation range of the former materials was substantially inferior to that of silver and gold due to the higher loss values of the former.
Chalcogenides for Photonic Applications

- Chalcogenides are materials containing elements of the sulphur group (sulphur, selenium, tellurium)
- They form glasses when combined with network formers such as As, Ge, Sb, Ga, Si or P
- These glasses have a transparency window in the spectral region from 2-25 µm
- They are often photosensitive, have high refractive indices \( (n \sim 2 - 3) \), high nonlinear refractive indices \( n_2 \), low Two Photon Absorption Coefficients (\( \beta \)), and good values for the non-linear parameter \( \gamma = \omega n_2 / (c A_{\text{eff}} \) \( A_{\text{eff}} \) being the mode area) and the Figure of Merit \( n_2 / \beta \lambda \).
### Table 1 | Nonlinear optical parameters for nonlinear waveguides used for all-optical signal processing.

<table>
<thead>
<tr>
<th>Device and material</th>
<th>$n_2$ (m$^2$ W$^{-1}$)</th>
<th>Nonlinear parameter $\gamma$ (W$^{-1}$ km$^{-1}$)</th>
<th>Reference</th>
<th>Dispersion coefficient (ps km$^{-1}$ nm$^{-1}$)</th>
<th>Loss (dB m$^{-1}$)</th>
<th>TPA (m W$^{-1}$)</th>
<th>FOM</th>
<th>Free carriers</th>
</tr>
</thead>
<tbody>
<tr>
<td>Highly nonlinear silica fibre</td>
<td>$3.2 \times 10^{-20}$</td>
<td>21</td>
<td>---</td>
<td>0.03</td>
<td>$10^{-3}$</td>
<td>Negligible</td>
<td>Large</td>
<td>No</td>
</tr>
<tr>
<td>Bismuth oxide fibre</td>
<td>$1.1 \times 10^{-18}$</td>
<td>1,360</td>
<td>100</td>
<td>$-260$</td>
<td>0.8</td>
<td>Negligible</td>
<td>Large</td>
<td>No</td>
</tr>
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<td>As$_2$S$_3$ fibre</td>
<td>$2 \times 10^{-18}$</td>
<td>160</td>
<td>101</td>
<td>410</td>
<td>0.88</td>
<td>$6.2 \times 10^{-15}$</td>
<td>208</td>
<td>No</td>
</tr>
<tr>
<td>As$_2$Se$_3$ fibre</td>
<td>$9 \times 10^{-16}$</td>
<td>1,200</td>
<td>102</td>
<td>$-504$</td>
<td>1</td>
<td>$2.5 \times 10^{-12}$</td>
<td>2.3</td>
<td>No</td>
</tr>
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<td>As$_2$S$_3$ rib waveguide</td>
<td>$2.9 \times 10^{-18}$</td>
<td>1,700</td>
<td>103</td>
<td>$-342$</td>
<td>5</td>
<td>$6.2 \times 10^{-15}$</td>
<td>304</td>
<td>No</td>
</tr>
<tr>
<td>As$_2$Se$_3$ fibre taper</td>
<td>$1.1 \times 10^{-17}$</td>
<td>93,400</td>
<td>99</td>
<td>282</td>
<td>$&lt;1$</td>
<td>$2.5 \times 10^{-12}$</td>
<td>2.84</td>
<td>No</td>
</tr>
<tr>
<td>As$_2$S$_3$ dispersion-engineered rib waveguide</td>
<td>$3 \times 10^{-18}$</td>
<td>9,900</td>
<td>92</td>
<td>29</td>
<td>60</td>
<td>$6.2 \times 10^{-15}$</td>
<td>312</td>
<td>No</td>
</tr>
<tr>
<td>Ge$<em>{0.7}$As$</em>{0.3}$Se$_{6.45}$ nanowire</td>
<td>$9 \times 10^{-18}$</td>
<td>136,000</td>
<td>68</td>
<td>70</td>
<td>250</td>
<td>$10^{-13}$</td>
<td>60</td>
<td>No</td>
</tr>
<tr>
<td>Photonic crystal Ag–As$_2$Se$_3$ waveguide</td>
<td>$7 \times 10^{-17}$</td>
<td>$26 \times 10^6$</td>
<td>104</td>
<td>Not provided</td>
<td>1,000</td>
<td>$&lt;4.1 \times 10^{-12}$</td>
<td>&gt;11</td>
<td>No</td>
</tr>
<tr>
<td>Silicon nanowire waveguide</td>
<td>$6 \times 10^{-18}$</td>
<td>$1.5 \times 10^5$</td>
<td>105</td>
<td>Engineered anomalous</td>
<td>400</td>
<td>$5 \times 10^{-12}$</td>
<td>0.77</td>
<td>Yes</td>
</tr>
</tbody>
</table>
How Much Can a Bare Thin Film Absorb?

- A bare thin film, structured or otherwise, can at most absorb 50% of incident light.
- If it does so, its reflection and transmission will both be 25%.
- This has been known for a while: Hadley and Dennison, JOSA, 1947 (unstructured), Botten et al PRB, 1997 (structured)
- Why the current interest?
- For example, photovoltaics- achieving photoabsorption in a very thin layer
Simple matter to achieve a thin film approaching the limit, with extraordinary properties!

Deposit a thin metal film on a non-wetting substrate; measure its resistance

Stop the deposition when the resistance starts to drop abruptly

Percolation threshold- isolated globules start to form connected network

Network requires point contacts- connected in both metal and dielectric phases

Metal thickness around 8-10 nm
Structured Thin Film Absorption-2

Fig. 1. Transmission electron micrograph showing the labyrinthine structure of a silver film in the region of the metal–insulator transition. The scale bar is 400 nm.
Structured Thin Film Absorption-3

Fig. 4. The spectral transmittance and reflectance of the film AG305 ($p=3 \times 10^{-1} \Omega \cdot \text{cm}$) are essentially constant in the infrared.

Fig. 7. The transmittance of AG30C in the far infrared. The noise is due to water absorbed by the KBr substrate. No strong wavelength dependence can be seen.
Total Absorption-1

- To go from 50% absorption to 100% absorption with a thin absorber, we need to couple it to a suitable reflector.
- An old idea- see Salisbury screen, first developed in the early 1940’s see (a), (b) below.

Figure 2. One-dimensional geometries for total absorption in a thin film, in order of increasing complexity. In each case, coherent interference causes cancellation of the shaded outgoing wave(s). (a) Geometry for a coherent perfect absorber, where both input beams are totally absorbed. (b) Critical coupling of a thin film with dielectric spacer and perfect mirror. (c) The film inside a Fabry–Perot cavity with partially transmitting front mirror ($|t| < 1$) and perfect back mirror.
Total Absorption-2

- The geometry in (a) needs two incident waves with the correct amplitudes and relative phase.
- In (b) and (c) there is only one incident wave.
- The condition for total absorption in (b) is $|r| = |r^2 - t^2|$
- The condition for (c) is that the transmission into the cavity is equal to the rate at which it absorbs energy: $|t|^2 = A$

\[\text{Figure 2. One-dimensional geometries for total absorption in a thin film, in order of increasing complexity. In each case, coherent interference causes cancellation of the shaded outgoing wave(s). (a) Geometry for a coherent perfect absorber, where both input beams are totally absorbed. (b) Critical coupling of a thin film with dielectric spacer and perfect mirror. (c) The film inside a Fabry–Perot cavity with partially transmitting front mirror (|H| < 1) and perfect back mirror.}\]
Fig. 1. (a) An ultra-thin layer in a symmetric background, illuminated from one side has $a \leq 0.5$. (b) The optimised absorber from (a), illuminated coherently from both sides has $a = 1$. (c) Halving the thickness of the absorber and placing it directly on a mirror is equivalent to (b). (d) A situation similar to (b) is created when a perfect conductor is placed behind the absorber at a spacing of $m_0 \lambda/4$. The ideal refractive index is somewhat different because the beam entering the absorber from below has passed through the absorber. (e) The top of a conductor is patterned with a surface grating allowing it to couple light into sideways propagating surface plasmons.

Fig. 2. Asymmetric Fabry-Perot etalons with perfectly conducting substrates. (a) Homogeneous (or homogenized) layer, (b) a Lamellar grating. Marked are the modal amplitudes and their reflection and transmission coefficients, which in (b) are contained in scattering matrices.
Total light absorption (TLA) condition for a homogeneous layer at normal incidence is

\[ r = -\gamma^2, \quad r = \frac{n - m}{n + m}, \quad \gamma = e^{ink_0h/2} \tag{1} \]

This is equivalent to critical coupling: loss rate of the resonance equals rate of incident energy.

The equation (1) can be derived as follows:

\[ \gamma r'\gamma b + \gamma ta = b, \quad \text{Fabry – Perot} \tag{2} \]

\[ t'\gamma b + ra = 0, \quad \text{Zero reflection} \tag{3} \]

\[ -rr' + tt' = 1 \quad \text{Energy conservation} \tag{4} \]
From (1) we can solve for the complex refractive index $n$ for TLA:

$$n = \frac{im}{\tan(nk_0 h/2)}$$  \hspace{1cm} (5)

For ultra-thin layers, expanding (5) gives

$$n \simeq \sqrt{\frac{im\lambda}{\pi h}} + \frac{1}{3}m^2$$  \hspace{1cm} (6)

Hence, if $\lambda/h >> m$, then $n' \simeq n''$, with $n''$ the slightly smaller. Also, $|n| \propto \sqrt{\lambda/h} - |n|$ only moderately large if $h$ is small.
Consider now TLA with thin lamellar gratings: $f$ denotes fraction of period $d$ which is $n$; the rest is air.

$d$ must be chosen so only specular orders exist outside the grating.

Require two Bloch modes to exist inside the grating: $n_{eff} d > \lambda$, where $n_{eff}$ comes from the linear mixing formula for permittivity for TE polarisation, and from the inverse law for TM.

The conditions for TLA become polarisation dependent, and the mechanisms for TLA are also polarisation dependent: TE– slab waveguide modes; TM– SPP’s.
Fig. 5. (a) Absorption as a function of complex refractive index for a 10 nm thick uniform film placed above an ideal conductor, illuminated by $\lambda = 700$ nm light. The contours have the same values as in Fig. 3. The thickness of the spacer layer between the absorbing layer and the reflector that maximizes the absorption. When $n = 1$ the spacer thickness is $\lambda/4$, which is marked by the blue curve.
The theory for two modes labelled 0 and 1 takes a similar form to the development in equations (2-4).

The Fabry-Perot equation for BoundMode1 (BM1) is

\[ \gamma_1 t_{01} a + \gamma_1 r_{01}' \gamma_0 c_0 + \gamma_1 r_{11}' \gamma_1 c_1 = c_1. \]  

(7)

The second term above comes from the BM0 giving a contribution to BM1 upon reflection at the top surface.

The condition (1) is now the vanishing of the determinant of a 3 by 3 matrix:

\[
\det \begin{bmatrix}
  r_{00} & t_{00}' \gamma_0 & t_{10}' \gamma_1 \\
  \gamma_0 t_{00} & \gamma_0 r_{00}' \gamma_0 - 1 & \gamma_1 r_{10}' \gamma_0 \\
  \gamma_1 t_{01} & \gamma_1 r_{01}' \gamma_0 & \gamma_1 r_{11}' \gamma_1 - 1 \\
\end{bmatrix} = 0.
\]  

(8)
Experiment-1

- The experiment was done at the ANU: Kylie Catchpole/ Tom White leading the group.
- The absorbing material - antimony sulphide Sb$_2$S$_3$: stable semiconductor; deposited by thermal evaporation; suitable complex $n$ for TLA near $\lambda = 600$nm
- Structure: 130 nm thick Ag substrate (thermally evaporated); 245 nm thick SiO$_2$ spacer layer (PECVD); 41 nm thick amorphous Sb$_2$S$_3$ layer deposited by thermal evaporation.
- Grating fabricated using EBL to define a mask in PMMA resist; ICP etch using CHF$_3$ gas.
- Two gratings fabricated to deliver TLA at 591 nm and 605 nm: $d = 375$ nm, $f = 72\%$; $d = 385$ nm, $f = 75\%$.
- At the two wavelengths: $n_{Sb_2S_3} = 3.342 + 0.096i$, $n_{Sb_2S_3} = 3.298 + 0.074i$
Figure 6: (a) Scanning electron micrograph at an angle of at 45° of the Sb$_2$S$_3$ grating structure with $d = 388$ nm etched groove width 97 nm (designed for TLA at $\lambda = 605$ nm). (b) Focused ion beam cut cross-sectional view where individual layers and the grating are clearly distinguished and labelled.
Figure 7: Reflectance of the fabricated gratings designed for TLA of TE polarized light at $\lambda = 591$ nm (green) and $\lambda = 605$ nm (red) and the planar reference structure (blue). The measured values (triangles, circles and squares respectively) and simulated predictions (dashed, dot-dashed and solid curves) show excellent agreement, in particular in the vicinity of reflectance minima. The inset shows the simulated absorption in the Ag reflector below the grating optimized for $\lambda = 591$ nm.
Final Comments: Chalogenides

- Total absorption polarisation dependent
- Could get round that with a doubly periodic grating structure
- Any weakly absorbing semiconductor could have been used in place of antimony sulphide $\text{Sb}_2\text{S}_3$.
- Still a narrow band solution; optimized for normal incidence
- Depending on the application, different solutions would give better angular and wavelength bandwidth
- The theoretical framework would be useful in such generalisations.
We are at an exciting juncture: improved microfabrication techniques, new materials, new designs coming together to open up new spectral windows and applications.

There are many developments going on I haven’t covered: e.g. topological structures, metasurfaces, nonlinear interactions, coupled wave systems, other governing equations.

In plasmonics, the problem of loss is leading to investigation of high index structures, surface waves, Fano resonances.

Many investigators rely on powerful simulation packages.

However, there will always be plenty of room for appropriate analytic approaches, and the insights they can deliver.
Thank your for your attention!
Biperiodic Chalcogenide Absorbers

E. Popov, A-L Fehrembach and R.C. McPhedran, Optics Express, 24, 16410 (2016)
# Biperiodic Chalcogenide Absorbers-2

Table 1. The parameters of the 10 optimised doubly periodic absorber systems: lattice type, constituents (1 for the matrix, 2 for the inclusion), spacer thickness, cylinder diameter, cylinder length, reflectance at the design wavelength and flux into the silver substrate. Distances in μm.

<table>
<thead>
<tr>
<th>System</th>
<th>Lattice</th>
<th>1</th>
<th>2</th>
<th>d</th>
<th>D</th>
<th>H</th>
<th>R</th>
<th>%Ref</th>
</tr>
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<tr>
<td>1</td>
<td>sqr</td>
<td>Sb₂S₃</td>
<td>air</td>
<td>0.252</td>
<td>0.247</td>
<td>0.04</td>
<td>3.5·10⁻³</td>
<td>2%</td>
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<tr>
<td>2</td>
<td>sqr</td>
<td>air</td>
<td>Sb₂S₃</td>
<td>0.220</td>
<td>0.308</td>
<td>0.04</td>
<td>6.5·10⁻³</td>
<td>2.5%</td>
</tr>
<tr>
<td>3</td>
<td>sqr</td>
<td>Sb₂S₃</td>
<td>SiO₂</td>
<td>0.192</td>
<td>0.343</td>
<td>0.04</td>
<td>4.8·10⁻³</td>
<td>6.2%</td>
</tr>
<tr>
<td>4</td>
<td>sqr</td>
<td>Sb₂S₃</td>
<td>SiO₂</td>
<td>0.287</td>
<td>0.280</td>
<td>0.04</td>
<td>2.1·10⁻³</td>
<td>2.0%</td>
</tr>
<tr>
<td>5</td>
<td>sqr</td>
<td>SiO₂</td>
<td>Sb₂S₃</td>
<td>0.234</td>
<td>0.322</td>
<td>0.04</td>
<td>3.4·10⁻³</td>
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<td>Sb₂S₃</td>
<td>air</td>
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<td>0.05</td>
<td>1.3·10⁻⁴</td>
<td>2.9%</td>
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Fig. 2. (Left) Reflectance (black, left scale) and absorptance (red, right scale) in the silver layer of the grating structure of Case 1, Table 1, as a function of wavelength. Full lines - exact values, dots - taking into account the single order (0,0) inside the SiO$_2$ homogeneous layer. (Right) Reflectance at the wavelength of minimum normal reflectance as a function of dimensionless direction cosines $\alpha$ and $\beta$. The electric field of the incident plane wave is along the $\beta$ axis.