# Plasmonic and biomimetic light-trapping for photovoltaics

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## ABSTRACT

The challenge when applying photonics to photovoltaics is the need to provide broadband, multiple-angle solutions to problems and both plasmonics and biomimetics offer broadband approaches to reducing reflection and enhancing light-trapping. Over millions of years nature has optimised nanostructures to create black, transparent, white and mirrored surfaces, the antireflective "moth-eye" structures are perhaps the best known of these biophotonic materials. In this paper we use simulated and experimental studies to illustrate how careful optimisation of nanoscale features is required to ensure the optimum match between reflectivity, spectral bandwidth and device quantum efficiencies. In the case of light-trapping by plasmonic scattering there is more room for design and specific spectral regions can be targeted by precise control of the size, shape and density of particular metal nanoparticles. We describe how the best opportunity for plasmonics within inorganic solar cells appears to be enhanced light-trapping of near-band edge photons.

**Keywords:** antireflection, light-trapping, plasmonics, biomimetics, photovoltaics, solar cells, nanophotonics

## 1. INTRODUCTION

For many photovoltaic devices and all silicon thin film devices efficient antireflection and light-trapping is very important, and increasingly these issues might be solved by nanophotonics. The mainstream photonics community is mostly concerned with rather narrow spectral bandwidths (usually emanating from lasers) and very efficient reflectors, waveguides and photonic bandgap materials can be manufactured by controlling refractive indices at a sub-wavelength (nanometre) scale. These photonic "devices" are extremely effective for around 100nm bandwidths, and can be broadened by employing multimode schemes or "chirping", but only with some loss of fidelity. Applying photonics to photovoltaics and spectral bandwidths of 800nm is a great challenge. Biophotonics and plasmonics are field of work that may offer solutions to broadband problems, in this paper we briefly review examples of how each might be used in photovoltaics.

## 2. BIOMIMETIC ANTIREFLECTION

### 1.1 Introduction

Reduction of optical reflection is important for many technologies it particularly important to photovoltaics. Traditionally within photovoltaics the approach has been to exploit destructive interference by application of single-layer antireflective coatings, however these can only reduce reflection to very low levels for the specific wavelength for which they are designed [1]. Increasingly broadband antireflection (AR) can be achieved with double or triple layer coatings but the availability of materials with appropriate refractive indices and problems with the matching of thermal expansion coefficients limit the applicability of these schemes. For very low reflection, thin film coatings are often combined with texturing at dimensions greater than the wavelength of light. For instance the inverted pyramid scheme found on high efficiency solar cells [2] is a highly effective scheme but is incompatible with thin film devices. Taking inspiration from the eyes and wings of some species of moth (Fig. 1a)[3-5], materials have been patterned on a scale below the wavelength of incident light to create biomimetic "moth-eye" AR surfaces [6].

A simple understanding of the moth-eye AR effect can be gained by considering incident light responding to a spatial average of the optical properties of any given interface volume dependent on the fraction of substrate to surrounding medium. The features on moth-eye surfaces are tapered and so this fraction gradually increases from the surrounding medium to the substrate, effectively blurring the interface and thereby reducing reflection across a relatively broad (400-

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900nm) spectral range. Eventually reflection will increase at long wavelengths when the interface thickness is small compared to the wavelength and at short wavelengths when structures are no longer sub-wavelength and are diffractive.

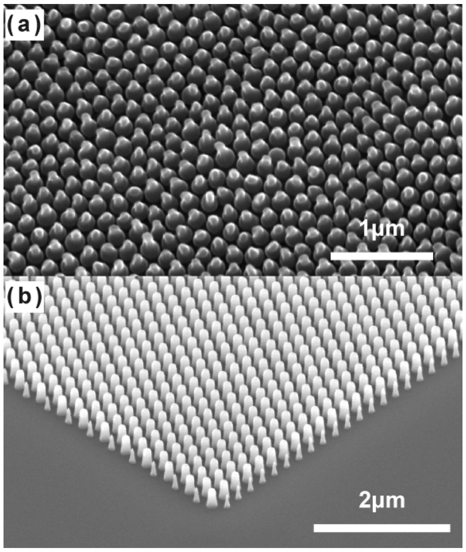


**Figure 1.** SEM images of (a) antireflective subwavelength features found on the transparent section of the wing of the *Cryptotympana Aquila*, (b) a biomimetic silicon moth-eye surface [6].

We have carried out detailed simulation and experimental studies to illustrate how this simple effective medium approach cannot accurately predict important features of moth-eye reflection spectra and shown that because the scale of moth-eye features is often only just below the wavelength of incident light, the height, shape, and inter-pillar spacing period of designs can all have a pronounced effect on optical properties and must therefore be considered for specific applications [6].

### 1.2 Experimental

In our studies electron beam lithography was used to define regular arrays on a silicon wafer. The period (pillar-to-pillar) distance of the pattern is varied from ~150 to ~350 nm, and the duty cycle (pillar width to period ratio) was varied from ~0.3 to ~0.7. A HBr anisotropic dry etch was used to transfer the resist pattern into the underlying silicon to create pillars with heights varying from ~150 to ~500 nm. AR surfaces were modelled using rigorous coupled wave analysis (RCWA) [7]. Results from our experiments show that a silicon moth-eye surfaces can exhibit extremely low reflectance (below 1% for wavelengths from 504 to 834 nm with a period of ~193 nm) (fig.2.). However, the reflectance spectra exhibit local maxima with spectral positions strongly dependent on the period of the array. Our simulations, based on similar nanostructure designs show similar spectral features and dependencies [6,8].

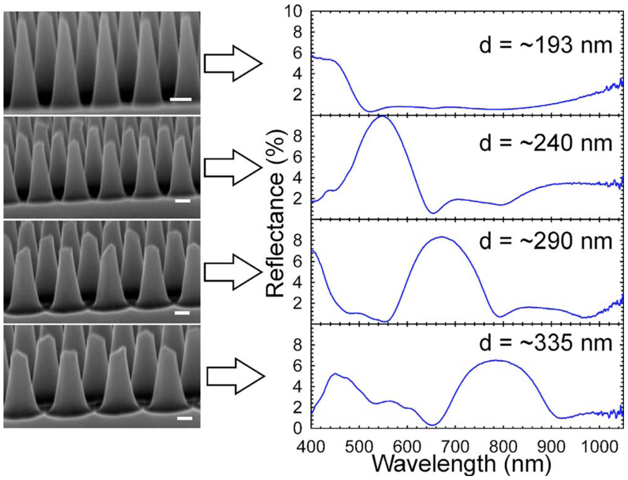


**Figure 2.** Measured reflectance spectra for silicon moth-eye surfaces of different periods. Scale bars in the SEM images are all 100 nm [6].

If reflectance of ideally shaped "moth-eye" arrays of 400nm height is plotted as a function of both periodicity and wavelength (fig.3), we can readily see that the best broadband low-reflectance for the solar spectrum (wavelengths range from 400nm to 1000nm) is obtained with periodicities in the 250nm to 300nm region.

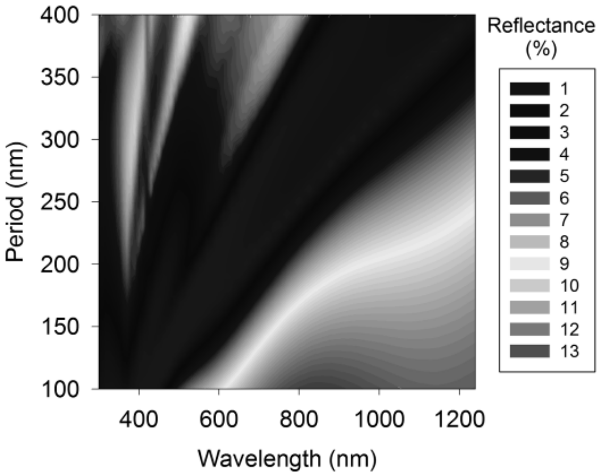


**Figure 3.** Variation of reflectance with the periodicity of a mimetic moth-eye array 400nm high as a function of wavelength. Extremely low reflectance's can be obtained for specific wavelength ranges [6].

If we take this reflectance data and consider the spectral data and typical silicon internal quantum efficiencies, we can plot expected maximum short circuit current density against periodicity. In our analysis we find that a periodicity of 280nm yields the highest values, though the current values seem to be relatively insensitive to large periodicity changes [8]. We find that the shape of the pillars can be much more significant and it is this feature that is often most difficult to control [6].

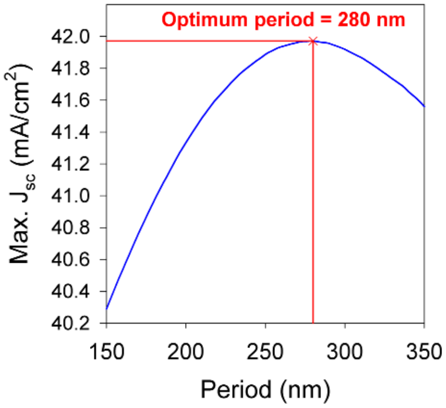


**Figure 4.** Consideration of spectral irradiance and internal quantum efficiency data indicates an optimum periodicity of around 280nm, the broadband nature of this type of surface is illustrated by the relative insensitivity to periodicity [8].

Optically, it is clear that carefully optimized moth-eye arrays could enhance solar cell performance, particularly compared to relatively narrow-band single and double layer antireflective coatings. However, it remains unclear that cheap and reliable large area techniques can be found that are cost competitive to pyramid texturing in the case of silicon wafer technology. The scheme is more likely to find application on thin film and concentrator devices, where it is possible to imagine that nanoscale surface texturing could be encouraged as part of the growth/fabrication cycle.

## 3. PLASMONIC LIGHT-TRAPPING

### 2.1 Introduction

A weakly absorbing material such as crystalline silicon (c-Si) requires thick layers and an extremely low defect and impurity densities to ensure high carrier lifetimes. Therefore solar cell grade c-Si is expensive to produce as it must be very high quality, and a large amount of material is required in each cell to sufficiently absorb the solar spectrum. Commercial c-Si solar cells are currently fabricated using wafers of around 200 µm thick. To reduce fabrication cost there is a trend towards ever thinner wafers, with predictions of 100 µm processes within the next five years, and less than 50 µm by 2020. In addition to reduced fabrication cost, thinner silicon layers have better electrical properties, as light is absorbed closer to the depletion region. However, extremely thin wafers present several production challenges (including stress-induced flexing and kerf losses), and less of the solar spectrum is absorbed by thinner layers, which leads to a decrease in device efficiency.

Light-trapping schemes can be used to alleviate this problem by increasing the effective optical path length within the device. Due to the high refractive index of silicon materials, once light is within the active layer it can be effectively trapped by total internal reflection, provided it does not impinge at normal incidence on the front surface. Therefore the effective optical path length can be increased by texturing the front and/or rear surfaces of a solar cell. Yablonovitch and Cody predicted a maximum path length enhancement factor of 50 for the case of a silicon slab with texturing on both surfaces [9]. Figure 5a illustrates the absorption of the AM1.5 spectrum by a 100 µm crystalline silicon layer (assuming zero reflection). Increasing the path length by a factor of 10 leads to a 7.2% increase in power absorption (Fig. 5b). The increase is more significant for thinner layers, with a 13.1% increase in power absorption for a tenfold path length increase in a 25 µm layer. The path length factor can also be considered as a materials reduction factor. A light-trapping method that increases the path length by a factor of 10 could enable a factor of 10 reduction of silicon usage while maintaining the same output power.

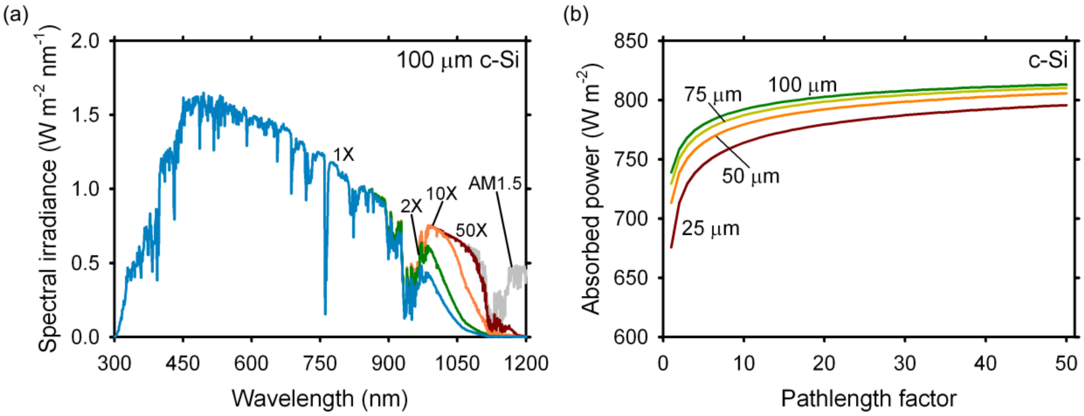


**Figure 5.** (a) Fraction of the AM1.5 spectrum absorbed by a 100 μm thick crystalline silicon layer assuming zero reflectance loss and ideal (lossless) light-trapping for path length factors of 1, 2, 10 and 50. (b) Corresponding absorbed power as a function of path length factor for four slab thicknesses.

Silicon can also be deposited at low cost onto glass and other substrates in the form of hydrogenated amorphous silicon (a-Si:H). Due to the disordered atomic structure of a-Si:H it features markedly different optical and electrical properties to c-Si. Optically, a-Si:H is a quasi-direct bandgap material and so has a much larger absorption coefficient than c-Si. However, the carrier diffusion length in a-Si is very small due to the high defect density. This small carrier diffusion length limits the layer thickness of a-Si:H solar cells to 300 nm or less. A 300 nm layer has moderate optical and electrical properties, decreasing the layer thickness will improve the electrical properties, but will also reduce the amount of light absorbed, increasing the thickness will absorb more light but lead to more carrier loss. Optimized devices find an appropriate compromise and modest efficiencies result. Unlike c-Si, the motivation to reduce a-Si:H layer thickness is mainly due to the improved carrier collection of thinner films. As with c-Si solar cells, light-trapping schemes enable reduction of layer thickness while maintaining sufficient absorption. Figure 6 demonstrates how a factor of ten increase in optical path length gives rise to a 27.6% increase in absorbed power for a 300 nm a-Si:H layer, and a 54.6% increase for a 100 nm layer. Even with light-trapping a-Si:H does not absorb as much of the solar spectrum as c-Si, as it has a higher energy bandgap (Fig. 6a). Other forms of thin-film silicon can be used in multi-junction devices to improve absorption of the solar spectrum, including microcrystalline Si (μc-Si:H) and microcrystalline SiGe (μc-Si$_{1-x}$Ge$_x$:H). Therefore there is a need to develop efficient thin-film light trapping for photons with wavelength up to ~1200 nm.

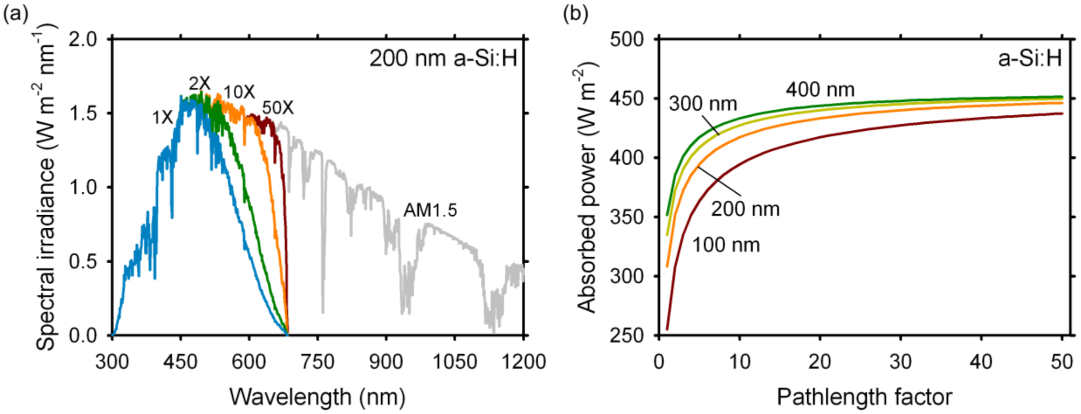


**Figure 6.** (a) Fraction of the AM1.5 spectrum absorbed by a 200 nm thick amorphous silicon layer assuming zero reflectance loss and ideal (lossless) light-trapping, for path length factors of 1, 2, 10 and 50. (b) Corresponding absorbed power as a function of path length factor for four slab thicknesses.

Most a-Si:H devices achieve light-trapping by using a textured surface that diffusely scatters light at an appropriate interface. Texturing of thin-film devices can be achieved by depositing onto a pre-textured substrate or superstrate. The film growth is conformal for thin layers, resulting in texturing of the front and rear surfaces. Most thin-film solar cells are deposited onto transparent conducting oxide (TCO) layers, which act as transparent top-contacts. TCOs can be textured during growth, or by plasma etching or wet chemical etching. An industry standard textured TCO is Type-U indium tin oxide (ITO) from Asahi [10]. However, although large improvements in cell efficiency are reported for devices fabricated onto textured substrates, the light-trapping achieved is far from optimum.

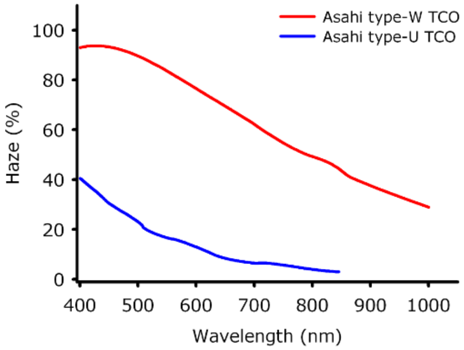


**Figure 7.** Haze measurements for Asahi type-W and type-U substrates [10].

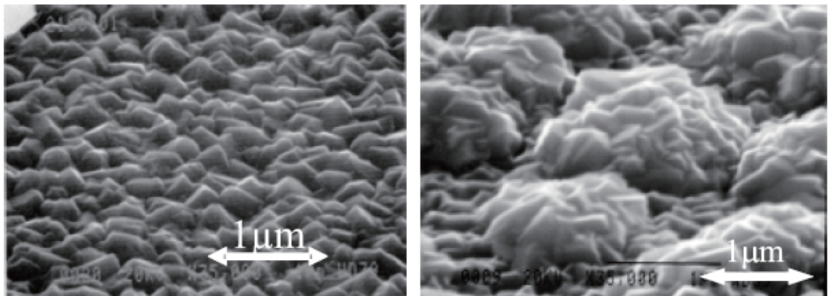


**Figure 8.** SEMs of roughened (left) Asahi type-U and (right) Asahi type-W substrates [10].

One measure of the effectiveness of a textured substrate is haze. Haze is defined as the ratio of transmitted light to diffusely transmitted light, *i.e.* 100% haze implies that all transmitted light has been diffusely scattered. Figure 7 shows the poor haze performance of Asahi Type-U, particularly in the NIR. Recently, Asahi developed a new version of textured ITO, Type-W, which offers considerably improved haze at longer wavelengths (Figs. 7,8)[10]. This improvement is due to increased surface roughness. In general, the surface texture must be on the scale of the wavelength of the photon we wish to scatter, and so large features are required to scatter IR photons. However, this roughness degrades the electrical properties of the film, as it does not provide a good surface for the layer to nucleate, resulting in poor film growth. Therefore there is a trade-off between improved light-absorption due to surface texturing, and degraded electrical properties due to poor film growth.

To reduce cost and improve the electrical properties of silicon films we require efficient light-trapping mechanisms that do not necessitate substantial roughening of the substrate. Many technologies developed by the photonics community may be of use for improving light-trapping and reducing reflection of solar cells. However, photonic experiments are typically concerned with only narrow emission ranges, and are fabricated without consideration of process cost. Therefore the critical challenges of applying photonic strategies to solar cells are the requirements of interaction over a broad range of wavelengths and angles of incidence, and the limit of a low process cost. Numerous photonic structures have been proposed for use in photovoltaic applications, including 2D diffraction gratings, distributed Bragg reflectors (DBRs) and photonic crystals. However, each of these structures either adds considerable process cost (*e.g.* requires the use of photolithography or electron-beam lithography), does not function well over a range of wavelengths, or both. In our view one of the most promising approaches to enhanced light-trapping involves the use of metal nanoparticles and utilization of localized surface plasmons (LSPs).

Investigation of metal nanoparticle enhancement of silicon solar cells is still at a very early stage, with just a small number of published experimental results [11-19]. So far, impressive gains in photocurrent at certain wavelengths are often accompanied by losses at other wavelengths and it is still not clear that metal nanoparticles can confer a net gain. The mechanisms responsible for photocurrent enhancement (or suppression) are not, as yet, well described in the literature. Over the last few years we have carried out a comprehensive study of these issues and have developed a detailed understanding of the opportunities for applying plasmonics to photovoltaics, in the remainder of this paper we summarize some of our findings [20].

### 2.2 Plasmonics

Metal nanoparticles, can exhibit an intense interaction with photons due to the excitation of localized surface plasmons. The wavelength range over which the interaction is strongest is highly tunable by modification of the particle size, shape and chemical composition, or by changes in the local dielectric environment. In addition to the potential for giant electric-field enhancement, metal nanoparticles feature extremely large optical cross-sections that often greatly exceed the respective geometric cross-section. In fact, metal nanoparticles interact with light more strongly than any other chromophore. For example, the absorption cross-section of Au nanoparticles is 4-5 orders of magnitude higher than strongly absorbing dyes such as indocyanine green, rhodamine-6G and malachite green [21]. The fact that the optical cross-section can greatly exceed the particle's geometrical area leads to the surprising realization that even sub-monolayers (*i.e.* monolayers with incomplete surface coverage) can scatter and/or absorb *all* incident photons across a broad range of wavelengths.

Unlike most other photonic structures, many of the broad range of nanoparticle fabrication and synthesis methods are well suited to covering large areas at low cost with minimal or moderate process complexity. The simplest approach is to moderately heat a thin continuous or semi-continuous film of metal, which results in the formation of a dense sub-monolayer of nanoparticles. The tools to achieve this are already in use as part of all standard a-Si:H and c-Si solar cell fabrication processes.

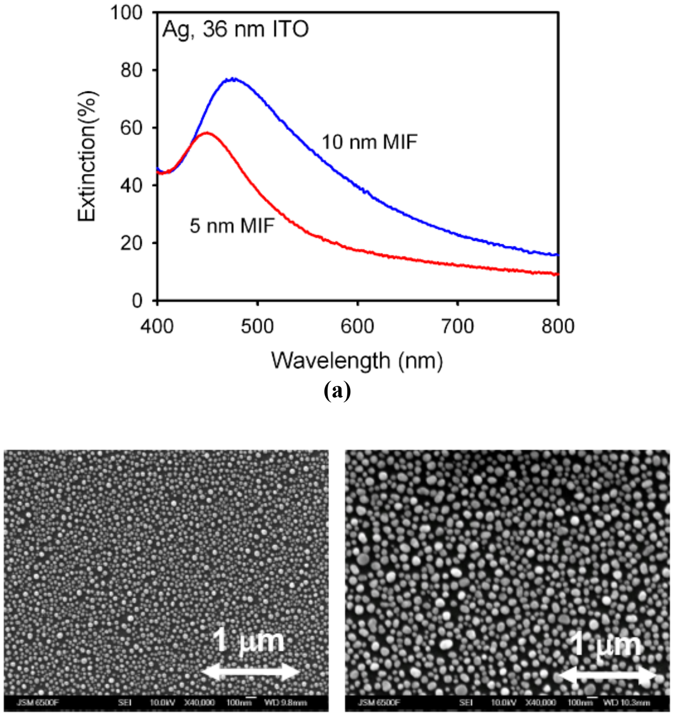
Metals can either absorb or scatter photons via plasmonic resonances that typically produce extinction peaks at around 400nm. Extinction is absorption plus scattering and the contributions and spectral distributions of each component can be modified by nanostructuring.

The scattering cross-section of some metal nanoparticles is exceptionally high and, combined with the ability to tune across the solar spectrum, this suggests that metal nanoparticles and scattering mechanisms can be used to improve light-trapping. Alternatively, plasmonic enhancement of local electric-field intensity could be used to enhance absorption in the active layer. Finally, if metal nanoparticles are able to inject excited carriers into the active layer, then absorption of photons by the nanoparticle is also of interest. However, as metal nanoparticles can also induce optical losses we must be careful to optimize their optical properties. These losses are a complex function of the nanoparticle size and shape, the chemical composition and the optical properties of the surrounding material.

Scattering represents the most immediate opportunity for thin film inorganic devices as we believe that absorption processes will generally be a cause of optical loss. In the case of solar cells based on organic materials, however, absorption may have a more significant role, as it should be possible to use the enhanced field strength of the localised plasmon to resonantly excite carriers in those systems since organic materials can afford to be in direct contact with the metal. So, when applying plasmonics to thin film silicon layers we need to pay attention to maximising scattering, preferably in the near-IR, while minimising absorption. By carefully considering the properties of the metal, the materials that surround the particles and the precise size and shape of particles, it is possible to optimise scattering behaviour.

### 2.2 Experimental

Distributions of metallic nanoparticles can be readily produced by annealing thin metal film, the extinction behaviour of two distributions of silver nanoparticles formed in this way are shown in figure 10. This self-organised technique produces a large size and shape distribution and in inhomogeneously broadened extinction features, but we can see that the larger particles shift the broad extinction peak towards longer wavelengths. In general we observe larger and sharper particles exhibit increasingly large extinction factors at longer wavelengths. Unfortunately, this type of metal island formation tends to produce broad extinction peaks that tend to have significant absorption contributions, particularly as a result of particle-particle interactions.



**(a)**

**(b)** **(c)**

**Figure 10.** (a) Extinction characteristics of silver nanoparticles formed by annealing (a) 5nm and (b) 10nm layers of silver. Extinction is sum of scattering (good for light-trapping) and absorption (a source of loss).

Although metal island films may provide an opportunity to illustrate enhanced light-trapping within some spectral regions of solar cells, there will always be a chance that losses will be greater than gains. It seems likely that carefully controlled densities of specific particle shapes and sizes that can maximize scattering at the near band edge region are most likely to provide the greatest benefits. With this in mind we have conducted simulations and experiments to determine the most promising forms of metal nanoparticles, in this regard silver and aluminum spheroids and nanorods appear particularly interesting.

Figure 11 illustrates the spectral distribution of extinction components for isolated silver spheroids (200nm long axis and 50nm short axis) as simulated using the separation of variables method of Voshchinnikov and Farafonov [22]. This simulation method allows us to readily calculate both the scattering and absorption components of extinction.

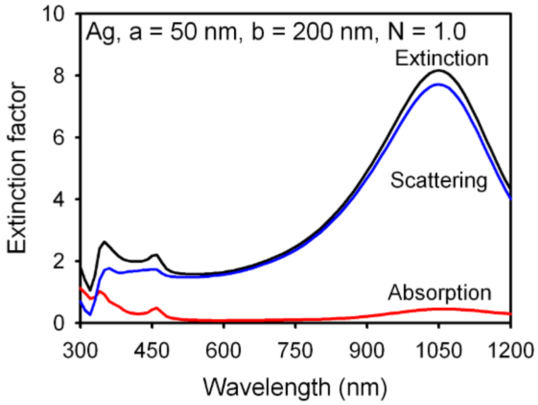
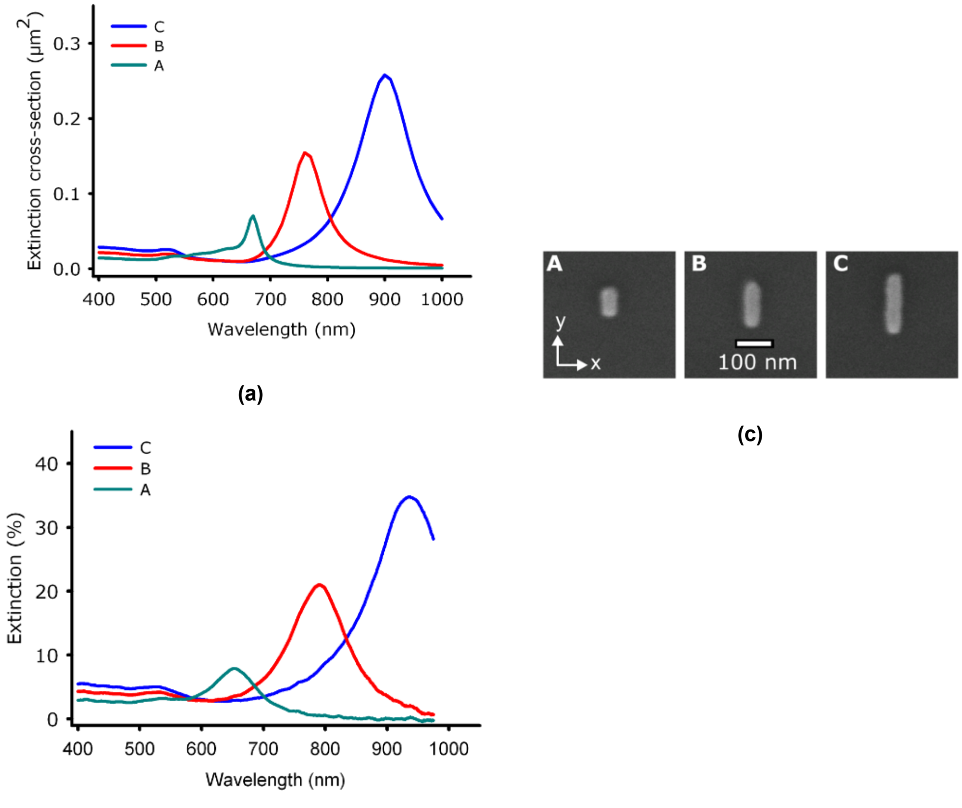


**Figure 11.** Simulated extinction, scattering and absorption spectra for silver spheroids

For silver spheroids we see that the extinction peak is shifted all the way to the silicon band edge and that this extinction is dominated by scattering. More extensive studies support the more general observation that larger particles with high aspect ratios and sharp angular features tend to have high extinction peaks that extend to long wavelengths and that the extinction in those regions are dominated by scattering. Absorption is relatively low across the whole spectral range but even small peaks at the short wavelengths and at the extinction peak would be causes for concern within a solar cell where photons are required to traverse the structure many times.



(a)

(c)

(b)

**Figure 12.** (a) Simulated and (b) Experimental extinction spectra for (c) three aluminum nanorods defined by e-beam lithography

With a view to maximising extinction (particularly the scattering component) in the near-IR we have carried out simulation and experimental studies of metal nanoparticles that could be defined by electron beam lithography. A number of metals, shapes and sizes have been studied [20] and we have found that nanorods, particularly aluminium nanorods are promising structures (Figure 12). Simulations here are now performed using discrete-dipole approximation (DDA) [23], and separating scattering and absorption components is computationally prohibitive. Meanwhile, in order to reduce costs, optical samples prepared by electron beam lithography are restricted to $1mm^2$ areas and this prevents an experimental determination of scattering/absorption ratios. For the aluminium nanorods we see massive extinction values (that we still expect to be dominated by scattering) in the near-IR. The 50nm by 200nm rods have the most promising properties with the highest extinction peak at a around 900nm.

These studies of electron beam defined metal nanostructures provide an indication of the types of structure that could be used to enhance light-trapping in the near bandedge region of inorganic solar cells. Of course electron beam lithography is an impractical fabrication method, a truly nanotechnological, preferably self-organising technique is required that will provide shapes, sizes and distribution densities of the most appropriate metal nanoparticles for the particular orientations and absorbing layers within devices.

## 3 CONCLUSIONS

Many properties of materials and solar cells are defined by features at a nanoscale. We are perhaps most familiar with the need to tune (spectrally match) semiconductor structures to optimize performance. In this paper we have illustrated parallels that exist within photonics and plasmonics concepts. In these instances careful selection and fabrication of features at a nanometer scale will determine appropriate spectral matching to areas of concern. In particular, we have shown that sub-wavelength A/R surfaces and plasmonic scattering materials can provide advantageous properties when carefully optimized for purpose.

Although, the moth-eye arrays can be readily understood by considering the nanostructures as an effective medium with a gradual variation of refractive index from air to semiconductor. In actuality the behavior of an array is more complex and features seen in the reflectance spectra cannot be explained by effective medium theory. We believe that moth-eye arrays can provide the best AR coating, however, enhanced device performance remains to be demonstrated. If moth-eye arrays can be shown to be advantageous, cheap and reliable large area techniques would need be found that are cost competitive when compared to other texturing or coating techniques. Moth-eyes are most likely to find application on thin film and concentrator devices, where natural pyramid texturing is not available and where it is possible to imagine that nanoscale surface texturing could be encouraged as part of the growth/fabrication cycle.

In plasmonics, it seems that arrays of very particular metal nanoparticles (such as aluminium nanorods) placed at above or below semiconductor layers within solar cells could provide greatly enhanced quantum efficiency in the near bandedge region. By carefully selecting the dimensions and distributions of the particles it should be possible to ensure that the peak of this plasmonic enhancement is spectrally matched to the near-IR. If this is done correctly the need for extremely rough “hazed” substrates should be reduced and this, in turn, should allow improved layer growth and better device performance. Alternatively enhanced light-trapping could allow thinner absorption layers for similar efficiency and cheaper devices. As with moth-eye antireflection, large area manufacturing techniques are required that can cheaply and reliably provide exactly the right nanoscale features, and relatively small deviations from the ideal could be very detrimental to device performance.

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