Embedded Ag@SiO₂ Nanoparticles for Enhanced Solar Absorption in Thin Film Photovoltaics

R.K. Harrison and Adela Ben-Yakar
Mechanical Engineering
University of Texas at Austin
Austin, Texas, U.S.A.

Abstract—we analytically determine the contribution of plasmonic nanospheres embedded in absorbing media to total optical absorption. We estimate gains of ~30% for a 1 μm μc-Si solar cell using 54 nm silica-coated silver nanoparticles.

Keywords—plasmonics; photovoltaics; light-trapping; nanoparticles

I. INTRODUCTION

Alternative light trapping techniques must be developed for thin film photovoltaics because conventional methods are not applicable to thin film applications. Plasmonics offers a potential solution for enhancing the optical absorption for thin film solar cells. Thin film solar cells have significant advantages over conventional ‘thick’ crystalline silicon cells in terms of the manufacturability and cost-to-power ratio, but thin film cells still lag in terms of overall efficiency. By integrating plasmonic particles tuned to selectively interact with weakly absorbed long wavelength light, the overall absorption and conversion of thin film solar cells can be enhanced. Conversely, fabrication costs and material demands can be reduced if the absorber thickness can be reduced at the same efficiency level. One proposed approach involves the incorporation of the plasmonic nanoparticle directly within the absorber material [1]. In this configuration, both enhanced near-field absorption and increased path length due to scattering by the particle will lead to increased absorption by the semiconductor. However, it is unclear which mechanism will be dominant and what parameters will dictate the magnitude of each effect. Here, we evaluate both contributions to the enhanced optical absorption in thin film photovoltaics.

Scattering in the perpendicular directions will provide a net gain in absorption within the cell for photons with energies greater than the bandgap, as the lateral dimensions of thin film solar cells are much larger than the thickness. Plasmonic particles are of significant interest because they result in strong high-angle scattering relative to dielectric particles, which scatter light mainly in the forward direction. The increase in absorption within the medium is calculated with ray tracing using complex Fresnel equations to determine reflection at the cell boundaries. By calculating light scattered outside of the near-field region, we avoid ‘double counting’ while still being able to account for all sources of absorption.

Silver core nanoparticles coated with a thin silica shell (Ag@SiO₂) are here considered for enhanced photovoltaic applications. Exposed metal surfaces within the active layer could drastically reduce charge collection efficiencies, so silica shells are proposed to mitigate charge trapping at the metal surface. Silver nanoparticles are examined because of the reduced cost, strong blue-shifted plasmonic response, and improved scattering-to-absorption ratio relative to gold spheres. Silica coatings allow for larger particles (and correspondingly higher scattering-to-absorption ratios) than would be normally feasible for a given material bandgap. It is necessary to keep the silica shell as thin as possible to avoid damping the plasmonic response. Experimental evidence indicates that an oxide thickness of ~2 nm is sufficient to effectively insulate a metal surface [2, 3]. Thus, we consider a 2 nm SiO₂ coating on a 25 nm silver core to yield a 54 nm overall particle size.

II. THEORY

Current simulation methods are insufficient for calculating plasmonic contributions to optical absorption. For example, screening many configurations with finite-difference time-domain codes is impractical. Furthermore, finite discretization of rounded particles with cubic domains can lead to errors in the enhancement, optical response and scattering behavior in the near-field. Therefore, there is a need for an exact analytical method to simulate the optical absorption around a plasmonic structure embedded in an absorbing material.

A. Extended Mie Theory for Spheres in Absorbing Media

We extend Mie theory equations for the electromagnetic fields at all points inside and around a coated sphere in an absorbing medium, building on previous published work for nanoparticles in absorptive embedding media [4] and solutions for coated spheres [5]. The spherical harmonic coefficients can be solved for using direct matrix inversion of the coated sphere boundary conditions. The fields can then be obtained using the same equations as an uncoated sphere in an absorbing medium.

B. Near-Field Absorption in an Absorbing Medium

For practical applications using plasmonic nanoparticles, we are interested in understanding how plasmonic near-field enhancement contributes to enhanced absorption in the surrounding medium. Here we have developed expressions for the deposition of energy in the near-field around spheres in an absorbing medium. Though the effect of an absorbing medium on the resonance of nanoparticles has been studied in some detail, rigorous studies of the absorption by the medium in the near-field are much less common. Several recent studies have recently addressed the problem of individual metal nanoparticles in absorbing media [6-8]. A control volume approach can be used to determine the total energy absorbed...
around a scatterer. Here we also determine the incident and scattered wave contributions to total energy deposition. Near-field optical localization and enhancement can increase the optical absorption, but a more important effect is the coupling of incident light into guided modes, where the absorption path length can be much larger than the thickness of the cell.

III. RESULTS

Fig. 1 shows the effect of a thin silica shell and an absorbing medium on the plasmonic response of a silver nanoparticle. We present the scattering efficiency (solid lines) and the scattering-to-extinction ratio (dotted lines, right axis). The configurations include a 50 nm Ag nanoparticles in: water, water with a 2 nm SiO₂ shell, microcrystalline silicon (μc-Si) medium, and μc-Si medium with 2 nm SiO₂ shell.

These results reveal a few points of interest. First, the silica shell results in only a slight red-shift in a water medium. For a μc-Si medium, however, a thin silica shell results in a dramatic blue-shift in the resonance. This behavior occurs because of the relative difference between the optical properties of the silver sphere and the shell relative to the surrounding medium. Interestingly, the presence of an absorbing medium also results in a significantly enhanced scattering-to-extinction ratio. We find that the extinction and absorption efficiencies decrease in the presence of absorption, but the scattering increases. This is a crucial factor for the use of plasmonics for solar applications, as absorption by the particle represents a parasitic loss.

Fig. 2 shows the net effects of Ag@SiO₂ nanoparticles embedded in μc-Si on the total optical absorption. We first determine the light intensity at the particle location, which is significantly modified from the incident solar spectrum. This is an important effect for the use of embedded nanoparticles, as significantly reduced fractions of light reach the particle in the short wavelength portion of the spectrum where the plasmonic gain is low. The light incident on the particle is then accounted for in terms of loss mechanisms, such as absorption by the particle, and gain mechanisms, such as near-field absorption and increased path length through scattering. We simulate a μc-Si solar cell in a superstrate configuration with a 1 μm absorber layer and an aluminum back contact. The particles are distributed in a close-packed hexagonal pattern at the midplane of the absorber layer spaced such that the total extinction cross sections do not overlap at any wavelength. We find that the near-field contribution dominates for strong absorbers such as CIGS, CdTe or amorphous silicon, but that the scattering mechanism is more important for weak absorbers such as μc-Si. Furthermore, the plasmonic enhancement is strongest when optical absorption is a limiting factor, such as weak absorbers and extremely thin solar cells.

Fig. 2 presents the incident solar spectrum, the spectral distribution reaching the particle, absorption by the silicon alone, conversion by the silicon alone, absorption by the silicon with embedded nanoparticles, and conversion by the silicon with embedded nanoparticles. A pronounced increase in the absorption is observed near the scattering peak of the nanoparticle plasmonic response. Efforts to experimentally validate these conclusions are ongoing.

REFERENCES