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Theory of random nanoparticle layers in photovoltaic devices applied to self-aggregated metal samples

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ABSTRACT

Random Al and Ag nanoparticle distributions are studied on varying substrates, where we exploit the nanosphere self-aggregation (NSA) method for fabrication. Relying on the measured particle size distributions of these samples, we develop a theoretical model that can be applied to arbitrary random nanostructure layers as is demonstrated for several distinct NSA samples. As a proof of concept, the optical properties of the exact same particles distributions, made from the quasi-random modelling input with electron beam lithography (EBL), are investigated from both theory and experiment. Our numerical procedure is based on rigorous solutions of Maxwell's equations and yields optical spectra of fully interacting randomly positioned nanoparticle arrays. These results constitute a new methodology for improving the optical performance of layers of nanoparticles with direct application to enhanced photovoltaics.

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1. Introduction

Metal nanoparticles (MNPs) possess promising optical properties, which improve light–matter interaction in a number of applications [1–5]. For example, these MNPs enhance light coupling in solar cells by means of efficient scattering [6–8] making them well suited to third generation photovoltaic devices. More generally, plasmon-assisted efficiency enhancement of solar cells may involve two classes of phenomena:

(a) For collective effects, a resonance condition determined by the geometry of the plasmonic structure and its environment is realized. Such resonances include localized surface plasmons (LSPR) and propagating surface plasmon polaritons (SPP). The former leads to high local field enhancement in the vicinity of the MNPs. Light driven processes such as multiple exciton generation [9,10] and photoluminescence [11] can be amplified as a result of this effect. Additionally, localized SPPs and propagating plasmons can be excited in waveguide structures within the solar cells [12].
(b) Scattering effects are observed for all frequencies and may be optimized over the whole optical spectrum, increasing the optical path length within the solar cell. Increased scattering and a high plasmon mode density enhances the effective absorptivity by efficiently coupling the light to the underlying structure [13]. These phenomena are particularly suitable for thin solar cell technologies [14].

Building on these ideas, the European FP7-248909-LIMA project [15] develops state-of-the-art techniques [11,16] in order to improve industry compatible light management in solar cells on the nanoscale. This is achieved by developing design rules of random plasmonic particle layer (PPL) specifications which may yield an overall increase in absorption efficiency. The PPL is combined with standard antireflection coating (ARC) technology, yielding improved performance and acting similar to cell texturing, as a result of light scattering at the PPL.

The solar cell design rules developed to date have concentrated on regular structures and optimized absorption efficiency for a narrow spectral range [17]. Random particle layers, however, are known to provide a broad spectrum of overlapping plasmon modes [18]. Light coupling efficiency increases with the number of modes that can be coupled into the system [19]. However, theoretical studies and predictions of the optical properties of random patterns are difficult [20,21]. Most research in this direction consider alloys or structured metal films [22,23] describing their optical properties with effective medium theory.

In this context, dielectric particles are as well investigated for the potential use of their photonic properties for photovoltaics [24,25].

In this paper, we use a simple route towards the modelling of optical spectra of random samples where only the particle size and distance distributions are needed as an input for optical calculations including interactions between these particles. We use this formalism to simulate the optical properties of specific random distributions of

Abbreviations: MNP, metal nanoparticle; LSPR, localized surface plasmon resonance; SPP, surface plasmon polariton; PPL, plasmonic particle layer; NSA, nanosphere self-aggregation (method); (R)EBL, (random) electron beam lithography; FTIR, Fourier transform infrared spectroscopy

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nanoparticles. Note that the theory of random distributions for photovoltaic applications can equally be applied for metal and dielectric structures. However, we concentrate on Al and Ag random layers.

In the following discussion, Section 2 describes the experimental industry-compatible nanosphere self-aggregation (NSA) method of PPL fabrication and characterisation. This yields experimental random distributions of particle geometries which are however difficult to model because of uncertainties in the actual spatial arrangement of the particles.

To resolve this issue, we present in Section 3 a combined experimental and theoretical study on the same exact particle distributions, chosen with a high degree of randomness. First, an exact quasi-random experimental distribution is constructed based on mimicking a truly random NSA distribution. This is achieved by electron beam lithography (EBL), following specific particle sizes and positions that are analytically defined. Then, the theoretical response is modelled for this quasi-random distribution and compared with the experimental data for the same exact distribution.

The combination of the analytically exact experiment and theory is a novel approach. It first allows validation of the modelling methodology by comparison with exact data presented in Section 3. This in turn allows the modelling of experimental self-aggregated particle distributions, yielding a methodology for optimizing experimental PPL properties.

NSA samples fabricated at both high and low temperatures are investigated. We discuss in detail our results in Section 4. Our conclusions derived from this work are given in Section 5. An overview on abbreviations used throughout this paper is provided above.

2. Nanosphere self-aggregation

A major challenge when exploiting plasmonic structures for solar cells is the production of nanoparticle layers with a cost-effective fabrication process. The nanosphere self-aggregation (NSA) method [26,27] provides a cheap, CMOS-compatible process, enabling integration of MNP layers in photovoltaic device manufacturing.

The NSA method, illustrated in Fig. 1(a), consists of annealing a thin Ag film which self-aggregates into a sheet of randomly distributed nanoparticles. The process parameters of the Ag precursor thickness, and annealing temperature and time, allow control of MNP geometrical parameters that influence particle homogeneity and density. To achieve a high degree of homogeneity, temperatures of hundreds of degrees Celsius are favored during the annealing process. The obtained particles are then hemi-spherical with negligible differences between minor and mayor axis. However, in our case we cannot use high temperatures in order to prevent damage of the solar cell, so that we restrict ourselves to the 100–400 °C temperature range, hence requiring 1–3 h of annealing. Finally, the choice of the Ag precursor thickness determines the mean particle size in the sample.

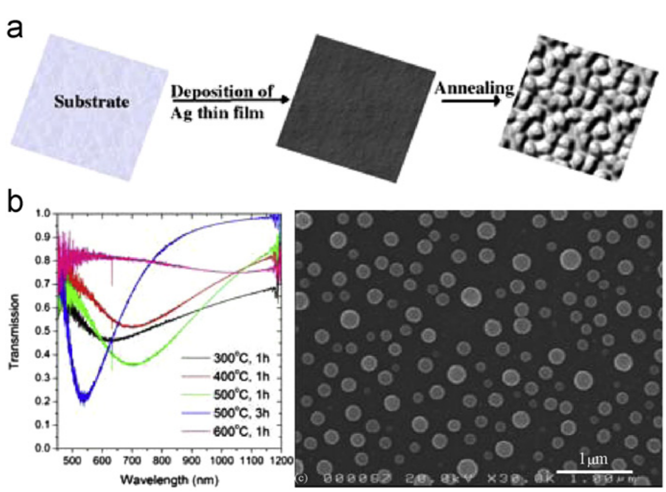

Fig. 1. (a) Schematic process of the nanosphere self-aggregation (NSA) method. (b) Optical characterization results for different annealing temperatures and times. This demonstrates how sensitive the optical properties are to fabrication parameters that result in different statistical characteristics of the NSA samples. (c) SEM image of a sample fabricated at 500 °C and annealed for 3 h for an initial Ag film thickness of 20 nm.

This method is well suited to fabricate large area films, and as such is suitable for mass fabrication of semiconductor solar cells. Other types of solar cells, such as synthetic dyes or organic polymer cells, are equally benefiting from the addition of plasmonic particles [28]. However, the MNPs need to be placed directly inside the dye or polymer, which requires methods such as electrodeposition or chemical deposition.

The resulting NSA samples are structurally characterised by scanning electron microscopy (SEM, Fig. 1(c)) and by spectrally resolved transmission and reflection measurements (FTIR). As can be seen from measurements under different anneal conditions in Fig. 1(b), the final particle layer characteristics crucially determine the optical spectra.

While the particle geometry obtained by NSA (Fig. 1(c)) is relatively homogeneous, the plasmon resonance is strongly dependent on average particle size, and the broadening is strongly related to the width of the particle size distribution.

The optical characterization demonstrates NSA plasmonic layer transmission (Fig. 1(b)) above 90% in the long wavelengths range, but also shows a reduced transmission for short wavelengths. This reduced transmission and associated loss due to reflection, interference, and absorption has been identified as a concern in prior art, as the losses at short wavelength range [27] compete with the enhanced absorption efficiency at long wavelengths. Solutions reducing the short wavelength loss include adjusting particle size and distribution, and incorporating an additional index matching layer on top of the PPL [29]. We numerically study the optical performance of random PPLs by applying exact electrodynamic modelling [30] summarised in the next section.

The resulting design methodology enables us to identify desirable particle diameters for the highest possible scattering efficiencies. The recommendations from the modelling are well within the wide range of parameters achievable with the NSA technique, and distributions with particle sizes of less than 100 nm in diameter are best suited. In this range, MNPs are efficient scatterers and dispersive losses are negligible [13,27]. A detailed image analysis provides us with information about the particle size and distance distribution. With the knowledge of these statistical parameters, we intend to predict optical spectra of related random patterns as obtained by the NSA method using the numerical procedure presented next.

3. Random e-beam lithography

Exact theoretical modelling of random PPLs is challenging. We introduce a quasi-random electron beam lithography (REBL) technique that enables direct comparison between experiment and modelling. EBL is an accurate nanofabrication method well suited for systematic study of precisely defined geometries, enabling exact comparison with modelling. Its resolution and the particle size in the quasi-random samples guarantee that aberrations from the ideal circular shape are negligible. In this section we therefore investigate exact quasi-random distributions theoretically and experimentally.

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