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journal homepage: [www.elsevier.com/locate/solmat](http://www.elsevier.com/locate/solmat)Light absorption enhancement for ultra-thin  $\text{Cu}(\text{In}_{1-x}\text{Ga}_x)\text{Se}_2$  solar cells using closely packed 2-D  $\text{SiO}_2$  nanosphere arraysG. Yin <sup>a,\*</sup>, P. Manley <sup>a</sup>, M. Schmid <sup>a,b</sup><sup>a</sup> Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Nanooptische Konzepte für die PV, Hahn-Meitner Platz 1, 14109 Berlin, Germany<sup>b</sup> Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany

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## ABSTRACT

2-D closely packed  $\text{SiO}_2$  nanosphere arrays serving as the photonic structure for light absorption enhancement on top of ultra-thin  $\text{Cu}(\text{In}_{1-x}\text{Ga}_x)\text{Se}_2$  solar cells are investigated both theoretically and experimentally. It is theoretically demonstrated that whispering gallery modes and high order Mie resonances contribute to the light absorption enhancement for the large spheres and an anti-reflection effect is prominent for small ones. The ultra-thin CIGSe solar cells achieve the optimum absorption enhancement for the small sphere array with a diameter of 110 nm, contrary to the larger spheres used in Si solar cells. The reason is attributed to the strong parasitic absorption in the AZO/ZnO/CdS front layers. They absorb mainly in the short wavelength range where the Mie resonances occur. Additionally, it is shown that the 110-nm-diameter sphere array exhibits a better angular tolerance than a conventional planar anti-reflection layer, which shows the potential as a promising anti-reflection structure.

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## 1. Introduction

To increase the competitiveness of  $\text{Cu}(\text{In}_{1-x}\text{Ga}_x)\text{Se}_2$  (CIGSe) solar cells and to obtain a large market deployment, further reduction of the cell manufacturing cost is desired. One approach to lower the cost is to reduce the thickness of the CIGSe photoactive layer, which allows for the reduction of material consumption, especially of the rare element In. However, reducing the thickness of the CIGSe photoactive layer from the typical 2–3  $\mu\text{m}$  to below 500 nm will inevitably lead to incomplete absorption of the incident light and will deteriorate the solar cell performance [1–3]. Therefore, light absorption enhancement is crucial to maintaining high efficiencies for ultra-thin (absorber thickness below to 500 nm) CIGSe solar cells. Previously, light absorption enhancement for CIGSe solar cells was achieved by coating an anti-reflection layer of  $\text{MgF}_2$  [4,5] or by improving the internal reflection at the CIGSe/Mo interface via inserting a dielectric layer [6] or via transferring the cells from the typical Mo back contact onto Au which has a better reflectivity [7]. Recently, a number of innovative nanoscale light-trapping structures have shown the potential to better improve the light absorption including plasmonic structures [8–11], dielectric diffractive nanostructures [12,13] and photonic crystals [14,15]. However, these innovative structures are mainly for Si-based, GaAs and organic solar cells, a

few have been reported for CIGSe solar cells but were limited to theoretical investigations [16–18]. The reasons for the difficulty of experimental implementation are assumed to be: Firstly, CIGSe solar cells are mostly deposited on Mo back contact, which makes the implementation of light-trapping nanostructures hard underneath the back contact; secondly, CIGSe absorbers are normally prepared at a substrate temperature above 500 °C and this temperature can trigger the diffusion of plasmonic materials (Au, Ag) prepared before CIGSe deposition.

The 2-D dielectric sphere array is a promising structure to be used as a mask for preparing nanostructures of other materials [19,20] and directly for light absorption enhancement in optoelectronic devices [15,21–23]. Confined resonant modes can be supported when the size of spheres is on the scale of the wavelength. The energy in the resonant modes can leak into the solar cells if the spheres are placed in close proximity [15]. Additionally, the dielectric sphere arrays have other advantages [15,19–24]: (1) dielectric materials are optically lossless, the concern of parasitic absorption is not necessary; (2) the geometry of the spheres is symmetric, which allows a broader acceptance angle of incident light; (3) the sphere array can be fabricated by the simple and cheap self-assembly method rather than complicated and expensive lithography technologies; (4) the inorganic dielectric materials are thermally stable compared to metallic materials, which makes them compatible to the deposition of the CIGSe layer at high substrate temperatures.

It has been implied that the 2-D colloidal  $\text{SiO}_2$  sphere array on top of a thin-film solar cell can significantly improve the light

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absorption, which has been demonstrated in amorphous Si solar cells [21,22]. However, since the structure and optical properties of CIGSe solar cells show a pronounced difference to Si solar cells, whether the 2-D colloidal SiO<sub>2</sub> sphere array can serve as an effective light-trapping structure for ultra-thin CIGSe solar cells is unknown. In this work, we investigate the optical influence of the SiO<sub>2</sub> sphere arrays on top of ultra-thin CIGSe solar cells and identify the underlying mechanisms of light absorption enhancement.

## 2. Simulations and experiments

To understand well the absorption enhancement provided by the 2-D SiO<sub>2</sub> nanosphere array on CIGSe solar cells, we performed 3-D simulations with the finite element method (FEM) using the software package JCMsuite [25]. JCMsuite is a commercial FEM solver specialized for nano-optical applications which solves the time harmonic form of Maxwell's equations. Fig. 1(a) illustrates an ultra-thin CIGSe solar cell with hexagonally closely packed SiO<sub>2</sub> spheres on top. The corresponding simulation cross section is presented in Fig. 1(b). The CIGSe solar cell had a typical structure of AZO(Al:ZnO)/ZnO/CdS/CIGSe/Mo from top to bottom. The corresponding thicknesses were 240/130/100/300/200 nm in the simulations presented here. A hexagonal computational domain with three sets of periodic boundary conditions in the  $x$ - $y$  plane and perfectly matched layer boundary conditions in the  $z$  direction were used. A plane wave source was used incident antiparallel to the  $z$  axis, in order to simulate light incident from above the solar cell. To avoid the calculation of point contact between spheres, an approximation of 5% overlap in diameter between neighboring spheres is assumed. Our simulations have proven that a slight overlap or distance between neighboring spheres will only lead to minor influence compared to the close case. The electric field intensity profiles ( $|E|^2/|E_0|^2$ ) in the following images were normalized to the incident electric field intensity ( $|E_0|^2$ ). To calculate the absorption in the layers of the solar cell, the total field volume integration inside those layers was used. The corresponding photocurrent was obtained by integrating the absorption in the absorber layer multiplied with the solar spectrum and assuming the complete conversion of absorbed photons to collected carriers under standard AM 1.5 illumination condition. To calculate the reflection, the integration of the Poynting flux of the scattered wave leaving the domain in the positive  $z$  direction was used. The optical constants of each layer were extracted from our own

samples via the transfer-matrix method [26]. The refractive index ( $n$ ) of SiO<sub>2</sub> was set to 1.46 [22]. We should mention here that only the absorption in the CIGSe photoactive layer ( $Abs_{CIGSe}$ ) can contribute to the photocurrent in the cell [27].

For experimental verifications of the reliability of the simulations, we prepared ultra-thin CIGSe solar cells. Because of the poor electric quality of solar cells with a 300-nm-thick CIGSe layer, the experimental CIGSe layer was 394 nm thick with a Ga/[Ga+In] ratio of 0.35. The thicknesses of the other layers were kept the same as in Fig. 1(b). For solar cell preparation details, please see [28].

The Langmuir–Blodgett method [24] is applied to prepare the 2-D closely packed SiO<sub>2</sub> sphere array due to its simplicity. The colloidal SiO<sub>2</sub> spheres used are commercially available and not surrounded by any chemical ligand. The array preparation starts with suspending the SiO<sub>2</sub> spheres in Butanol, then the SiO<sub>2</sub> suspension is dropped onto the surface of water, in which the solar cells are submerged in a petri dish. SiO<sub>2</sub> spheres spread on the water surface and form a monolayer after Butanol evaporates. Water in the petri dish was subsequently sucked off via an injector until the water level was below the surface of the solar cells. Finally, the result is a 2-D SiO<sub>2</sub> closely packed sphere array on the top of solar cell. In the whole preparation process, the water is the principal liquid medium used and no extra stabilizing agent is needed, which is compatible with the stability of CIGSe solar cells. The area of the petri dish was 5\*5 cm<sup>2</sup>, but this technology can be easily scaled up to module dimensions.

Scanning electron microscopy (SEM) is used for the characterization of the morphology of SiO<sub>2</sub> nanosphere arrays on top of solar cells. The current density–voltage ( $J$ - $V$ ) curves were measured under standard test conditions (AM 1.5, 100 mW/cm<sup>2</sup>, 25 °C) by a home-made system with a sun-simulator consisting of both a Xenon and a Halogen lamp. The AM1.5 condition is calibrated by a certified crystalline Si solar cell. The external quantum efficiency (EQE) was measured with a two-source illumination system of a Xenon and a Halogen lamp, using calibrated Si and Ge diode as references.

## 3. Results and discussion

### 3.1. Large sphere array

#### 3.1.1. Whispering gallery modes (WGMs)

Fig. 2(a) represents the simulated absorption in the CIGSe layer ( $Abs_{CIGSe}$ ) without and with 600-nm-diameter and 110-nm-

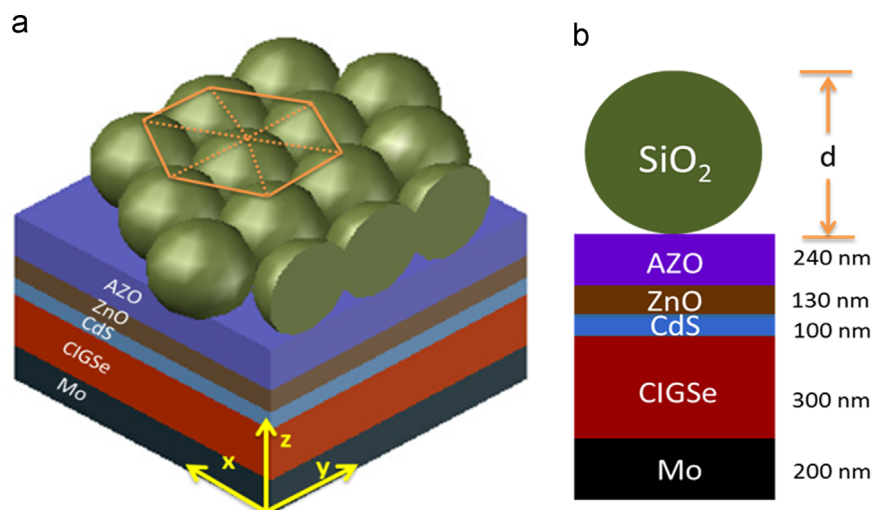


Fig. 1. (a) Schematic illustration of hexagonally closely packed SiO<sub>2</sub> spheres on top of an ultra-thin CIGSe solar cell, and (b) corresponding cross section of a single sphere on top.

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