

Integration of plasmonic Ag nanoparticles as a back reflector in ultra-thin Cu(In,Ga)Se₂ solar cells

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Abstract: Integration of plasmonic Ag nanoparticles as a back reflector in ultra-thin Cu(In,Ga)Se₂ (CIGSe) solar cells is investigated. X-ray Photoelectron Spectroscopy results show that Ag nanoparticles underneath a Sn:In₂O₃ back contact could not be thermally passivated even at a low substrate temperature of 440 °C during CIGSe deposition. It is shown that a 50 nm thick Al₂O₃ film prepared by Atomic Layer Deposition is able to block the diffusion of Ag, clearing the thermal obstacle in utilizing Ag nanoparticles as a back reflector in ultra-thin CIGSe solar cells. Via 3-D finite element optical simulation, it is proved that the Ag nanoparticles show the potential to contribute the effective absorption in CIGSe solar cells.

Keywords: ultra-thin Cu(In,Ga)Se₂ solar cells, Ag nanoparticles, thermal stability, Al₂O₃ film, atomic layer deposition

1. Introduction

In the last decade, tremendous attention has been paid to Cu(In,Ga)Se₂ (CIGSe) solar cells with thinner absorbers [1-6], which enables the reduction of consumption of rare material indium (In) and resulting manufacturing cost compared to their thick counterparts. However, it is demonstrated that high efficiencies (> 15 %) can only be maintained when the CIGSe absorber is thicker than 1 μm [1, 3, 7]. One of the dominant reasons is the incomplete absorption of incident light arising from the absorber thickness reduction [1-5], which leads to a much lower current density. Our final goal is to obtain highly efficient ultra-thin CIGSe solar cells (with CIGSe

absorber thickness below 500 nm). Therefore, light trapping is crucial to maintaining high efficiencies for ultra-thin CIGSe solar cells. Among various light-trapping technologies, subwavelength plasmonic metallic nanostructures have shown pronounced light-trapping effects in thin-film solar cells [8-11], because they can exhibit localized surface plasmons and increase the absorption of solar cells either by strong scattering or local near-field concentration.

The CIGSe solar cell has a typical structure of Al:ZnO(AZO)/i-ZnO/CdS/CIGSe/Back contact/glass substrate from top to bottom. Regarding the location of nanoparticles in the solar cell, we can in principle place the nanoparticles at any interface. For the consideration of favourable electrical properties of solar cells, it is not advisable to place the metallic nanoparticles at any of the interfaces within the AZO/ZnO/CdS/CIGSe/back contact layers. To avoid parasitic absorption and lower transmission originating from the Fano effect [12-14] at wavelengths below the surface plasmon resonance, placing the particles on the top of solar cells (air/AZO) is not recommended either. In this work, therefore, we limit our investigation to the configuration of placing metallic nanoparticles at the rear interface of back contact/glass substrate as a back reflector. Since the decaying length of near field is only a few nm, the enhanced near fields surrounding the particles will not penetrate into the absorbing layer. This indicates that only scattering from the particles will contribute as the light-trapping mechanism in this configuration. The concept of metallic nanoparticles as a back reflector has been both experimentally and theoretically observed in thin-film Si solar cells [9, 15-17]. However, there have been few reports of CIGSe solar cells using metallic nanoparticles as a back reflector. Compared to amorphous Si solar cells, the experimental challenges of incorporating metallic nanoparticles into CIGSe solar cells are as follows: 1) the deposition of amorphous Si solar cells can be done at a low substrate temperature around 250 °C or even lower [18, 19], which is not high enough to trigger the diffusion of metallic particles underneath the back contact. In contrast, the CIGSe absorber is normally deposited at above 500 °C [20], which poses a high risk of triggering the diffusion of metallic material through the back contact into the absorber. 2) The conventional back contact layer of CIGSe solar cells is the opaque Mo rather than transparent conductive oxide (TCO), which makes it hard to harvest the light-trapping benefit from the rear side.

TCO substrates have shown the potential to be promising alternative back contacts for CIGSe solar cells [21, 22]. Further, in our previous work [5], it was demonstrated that low substrate temperature (440 °C) could also enable efficient ultra-thin CIGSe solar cells. The low substrate

temperature makes it possible that metallic nanoparticles can be thermally passivated by a TCO back contact. The two points provide possibilities to utilize metallic nanoparticles as light-trapping structures at the rear interface (back contact/glass substrate) of CIGSe solar cells. Thus, in this work, we will investigate the potential of using metallic nanoparticles as light-trapping structures in ultra-thin CIGSe solar cells deposited at low substrate temperature on TCO substrate.

2 Experiments and characterization

Ag nanoparticles: Ag is selected as the plasmonic material in this work because of its strong scattering ability and low parasitic absorption [23]. The Ag nanoparticles were prepared by the so-called surface-tension-induced agglomeration method [15, 24]. A 30 nm thick Ag film was first grown on a glass substrate by thermal evaporation and was then annealed at 450 °C for 30 minutes in ambient atmosphere.

Al₂O₃ thin films: An Al₂O₃ thin film was introduced to thermally passivate the Ag particles before TCO deposition. The Al₂O₃ films were prepared by Atom Layer Deposition (ALD) using a BENEQ TFS200 ALD system. Trimethyl aluminium (TMA) and oxygen plasma were used as precursors for the plasma-enhanced ALD process at room temperature [25].

Solar cells: For the device completion, a 200 nm thick Sn:In₂O₃ (ITO) layer was sputtered on the prepared Ag particles as the TCO back contact. Subsequently, a 460 nm thick CIGSe absorber was fabricated by 3-stage co-evaporation process at a low substrate temperature of 440 °C. The [Ga]/[Ga+In] and [Cu]/[Ga+In] ratio are 0.35±0.01 and 0.87±0.01, respectively. A 50 nm thick CdS layer was then grown by chemical bath deposition (CBD). A sputtered 130 nm i-ZnO and a 240 nm Al doped ZnO (AZO) layer followed. The Ni/Al front contact was evaporated through a shadow mask with a total thickness of 2.5 µm. Finally, the solar cells were mechanically scribed to 0.5 cm² for a single solar cell. For comparison, solar cells without the incorporation of Ag nanoparticles were prepared simultaneously.

Characterization: For characterization of morphologies, scanning electron microscopy (SEM) was used. To investigate the thermal stability of Ag nanoparticles during CIGSe deposition, X-ray photoelectron spectroscopy (XPS) was applied. Transmittance (T) and reflectance (R) measurements of the samples under normal incidence were carried out by UV-Vis spectrometer with an integrating sphere, Absorption (Abs = 1-R-T) was deduced. To theoretically demonstrate

the light-trapping effect of Ag nanoparticles, 3-D numerical simulations were carried out using a Finite Element software package (JCMsuite) [26]. The simulation unit cell consists of the basic structure of AZO/ZnO/CdS/CIGSe/ITO/Al₂O₃/glass substrate with the thicknesses of 240/130/100/460/200/50/200 nm from top to bottom. The structure and thickness values were chosen according to the experimental samples. The Ag nanoparticles are placed at the rear interface of ITO/glass substrate. The unit cell is a hexagonal prism, consisting of three sets of periodic boundary conditions in the x-y plane. Perfectly matched layers (PML) are used in the both of the z directions to remove artificial reflections. Since the glass substrate block is too thick to be included inside the FEM domain, only 200 nm of glass substrate is included in the computational domain. Since we have a PML layer, it means that reflections from the rear glass side are neglected.

3 Results and discussion

3.1 Ag nanoparticles

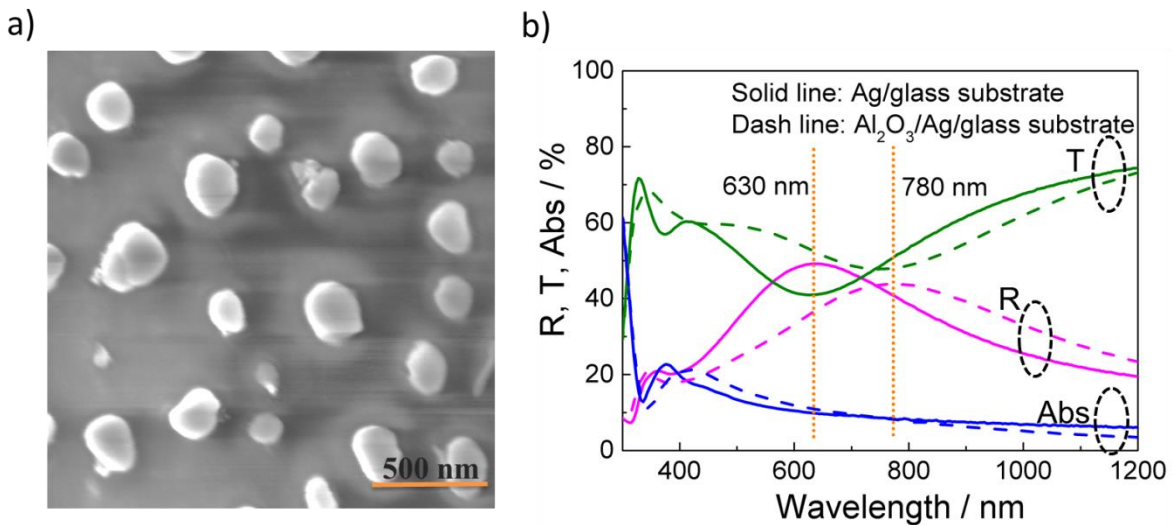


Fig.1 a) Surface morphology, b) reflectance (R), transmittance (T), absorption (Abs) of Ag nanoparticles on glass substrate

Fig.1 a) shows the surface morphology of Ag nanoparticles prepared by surface-tension-induced agglomeration method. We can observe approximately spherical Ag particles. They are randomly separated and particles of 200 nm major diameter dominate the size distribution. The corresponding optical responses R/T/Abs ($Abs = 1 - R - T$) were measured and are depicted in Fig.1

(b) (solid lines). There is a broad resonance peak centered at the wavelength of 630 nm. This corresponds to the dipole resonance of the Ag nanoparticles. The broadness of the resonance peak is due to the non-uniform size distribution and is actually beneficial for the broadband light-trapping requirement. Besides, the absorption for the prepared nanoparticles is below 10 % beyond 600 nm, which indicates that the scattering predominates over parasitic absorption over most of the wavelength range of interest. As to the parasitic absorption at wavelengths below 600 nm, this is not deleterious to the light-trapping effect since light in this wavelength range is mainly absorbed by the solar cell before reaching the Ag nanoparticles at the interface of ITO/glass substrate.

3.2 Thermal stability of Ag nanoparticles

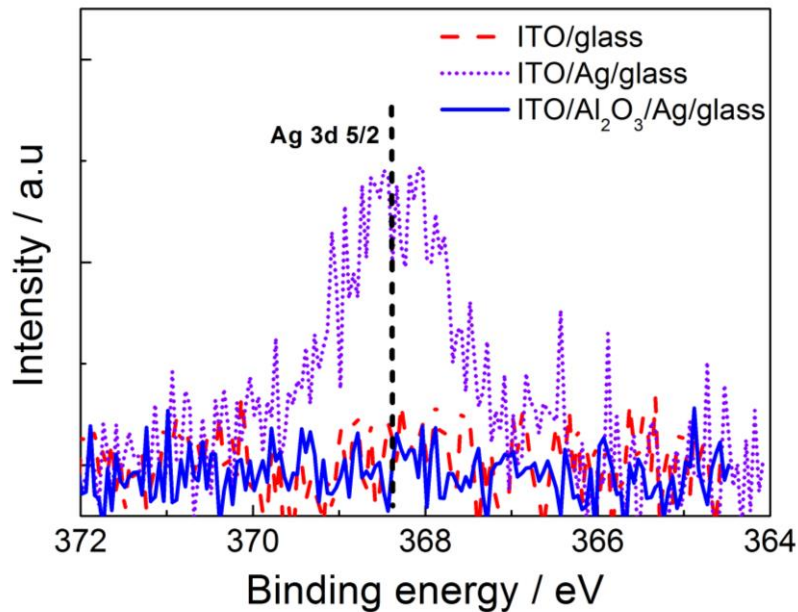


Fig.2 Investigation of Ag diffusion after the CIGSe deposition

Although the CIGSe deposition temperature was largely reduced from standard high temperature (above 500 °C) to 440 °C, we still could not ensure the thermal passivation of Ag nanoparticles. We investigated the sample of Ag nanoparticles coated by a 200 nm thick ITO back contact layer (ITO/Ag/glass substrate), which experienced the whole CIGSe deposition process but was blocked from CIGSe deposition by a mask on top. XPS was used to characterize the ITO surface and the result (shot dotted line) is shown in Fig.2. The signal of Ag was still detected, which indicated that the ITO layer failed to thermally passivate the Ag nanoparticles even at the reduced

substrate temperature during the CIGSe deposition. Further lowering the CIGSe deposition temperature may help block the diffusion of Ag, this will however lead to low-quality CIGSe absorbers and is not recommended. Increasing the thickness of the ITO layer is another alternative approach, this is however not feasible either. Because ITO has strong absorption due to free charge carriers, especially in the near infrared range, thicker ITO layers imply that more of the light scattered back from Ag nanoparticles will be dissipated in ITO. A third approach is to insert a passivation layer between ITO and Ag nanoparticles. This passivation layer should fulfill at least two requirements: 1) there is no absorption in the wavelength range of 600-1200 nm, where ultra-thin CIGSe solar cells have poor absorption; 2) this passivation layer should be highly compact and thermally stable. Accordingly, dielectric materials like Al_2O_3 and Si_3N_4 are materials of choice. Considering the available experimental conditions, Al_2O_3 is selected in this work.

The Al_2O_3 films were prepared by plasma-enhanced ALD at room temperature. Fig.2 shows the XPS result of the sample (solid line) with a 50 nm thick Al_2O_3 film. It can be observed that the 50 nm thick Al_2O_3 film was able to block the diffusion of Ag. We should stress here that whether the Al_2O_3 film can passivate the diffusion of Ag is dependent on how the Al_2O_3 film is prepared. We also tested a sputtered 150 nm thick Al_2O_3 film, which failed to block the diffusion of Ag (not shown here). This is due to the fact that the ALD-prepared Al_2O_3 film has a better conformity to the surface features and is much more compact than the sputtered.

Fig.3 shows the cross section and the surface topography of the sample of ITO/ Al_2O_3 /Ag /glass substrate. The cross section shows that the Ag nanoparticles are conformally covered by the Al_2O_3 film. Moreover, the ITO layer is also laterally continuous on top of the Al_2O_3 layer and no fracture is observed, which is critical for electrical properties of solar cells. The top view also confirms the conformal growth: the Ag nanoparticles tend to 'grow' bigger and become closer.

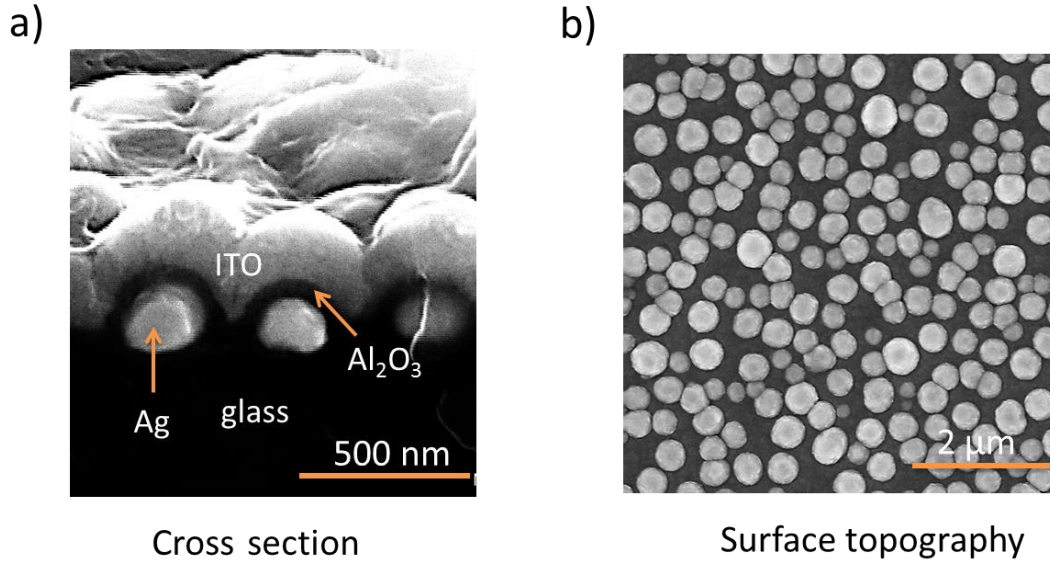


Fig.3 Morphologies of the structure of ITO/Al₂O₃/Ag/glass substrate

The Al₂O₃ coating can influence the optical responses of Ag nanoparticles as well. Fig.1 also shows the optical responses of Ag nanoparticles after the 50 nm thick ALD-prepared Al₂O₃ film was coated (dash lines). Compared to the optical responses without the Al₂O₃ film, the main feature is that the dipole resonance peak red-shifts from the wavelength 630 nm to around 790 nm due to the higher refractive index of Al₂O₃ than air [8, 15]. This redshift is actually beneficial for light trapping since the resonance peak is located at the center of the poorly absorbed wavelength range (600 - 1200 nm) for ultra-thin CIGSe solar cells.

3.3 Integration of Ag nanoparticles in solar cells

To confirm the light-trapping effect of Ag nanoparticles, the solar cells were prepared on top of ITO/Al₂O₃/Ag/glass substrate and ITO/Al₂O₃/glass substrate. Unfortunately, the cells with Ag nanoparticles showed poor electric performance. The reason is assumed to be related to the texture of the ITO layer as a result of the conformal growth as shown in Fig.3. Each single solar cell was mechanically scribed from 2.5*2.5 cm substrate to 0.5*1 cm. However, ITO has surface texture due to the conformal growth on Ag nanoparticles (see Fig. 3) and the distance between the neighbouring two nanoparticles is less than the size of the mechanical needle. Therefore, the mechanical needle either failed to completely separate the CIGSe absorber or cut off the ITO layer completely. This can shunt or open circuit the cells.

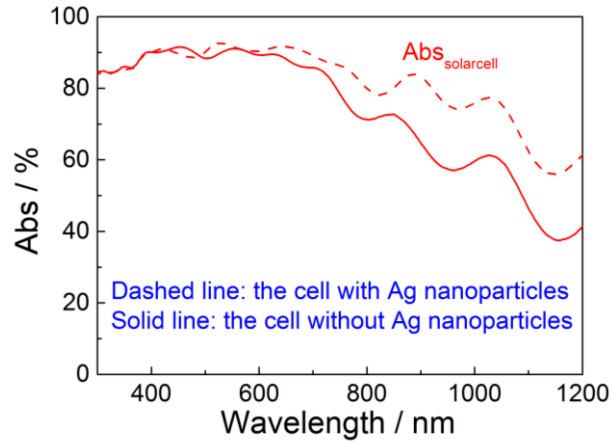


Fig.4 Absorption of the complete cells ($Abs_{solarcell}$) with and without Ag incorporation

R and T of the complete solar cell with and without Ag nanoparticle incorporation were measured and the absorption of the complete solar cell ($Abs_{solarcell}$) was calculated (in Fig.4). $Abs_{solarcell}$ is greatly enhanced in the wavelength range of 600-1200 nm due to the presence of the Ag nanoparticles. Since the solar cells showed poor electric performance, it is difficult to identify whether the increase in $Abs_{solarcell}$ is due to the effective absorption in CIGSe (Abs_{CIGSe}) or to the parasitic absorption in either Ag nanoparticles or/and any of the other layers.

To distinguish this point, 3-D simulations were carried out to check whether Ag nanoparticles can contribute to the effective Abs_{CIGSe} using the FEM software package (JCMsuite). The simulation structure is illustrated as Fig.5 a). To approximate the effect of an ensemble of different particle sizes, we simulate only the mean particle size. This was experimentally determined to be a hemispherical shape with a height of 40 nm, a width of 150 nm and a radius of curvature of 90 nm. This structure of $Al_2O_3/Ag/glass$ substrate in the simulation gives a comparable resonance (around 800 nm) to the experimental sample as shown in Fig.2. b) Conformal growth of layers on top of Ag nanoparticles is not taken into account. Since we only intend to confirm whether Ag nanoparticles are able to contribute to Abs_{CIGSe} , the simulations are sufficient to confirm the feasibility of this mechanism. Fig.5 b) shows both the calculated Abs_{CIGSe} and $Abs_{solarcell}$ with and without Ag nanoparticles. After incorporating Ag nanoparticles, the $Abs_{solarcell}$ is generally higher in the poor-absorbing spectrum region, which agrees with the changing trend of the experimentally measured in Fig.4. Similar to the $Abs_{solarcell}$, Abs_{CIGSe} is improved as well, which indicates that Ag nanoparticles are able to improve the effective Abs_{CIGSe} . Further, from the comparison of differences between $Abs_{solarcell}$ and Abs_{CIGSe} for solar cells with and without Ag

nanoparticles, the improvement of Abs_{CIGSe} is the dominant part of the increase of $Abs_{solarcell}$. Therefore, we can conclude that Ag nanoparticles can improve the effective absorption in CIGSe layers. Here, it is stressed again that the goal of this work is to prove the concept of light-trapping effects of Ag nanoparticles at the interface of ITO/glass substrate. For the maximum absorption enhancement, an optimization of particle shape, size and density should be performed.

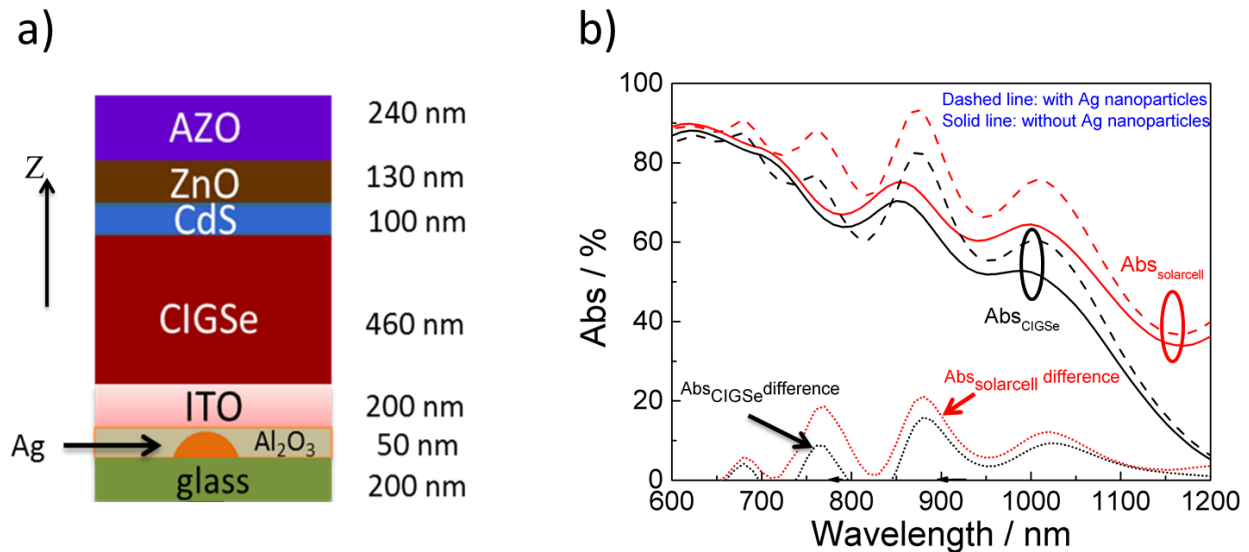


Fig.5 a) Cross section of the unit cell in the finite element method (FEM) simulation; b) simulation of Abs_{CIGSe} and $Abs_{solarcell}$ with and without Ag nanoparticles at the interface of ITO/glass substrate

4 Conclusion and outlook

In this work, incorporating Ag nanoparticles at the ITO/glass substrate interface as a back reflector in CIGSe solar cells has been investigated. It is confirmed that Ag nanoparticles can still penetrate the ITO back contact even at a low substrate temperature of 440 °C. The ALD-prepared Al₂O₃ film with a thickness of 50 nm is proved to be able to thermally passivate the diffusion of Ag. This addressed the thermal obstacle to utilize the Ag nanoparticles as a back reflector in CIGSe solar cells. With the help of the optical 3-D FEM simulation, it was proved that Ag nanoparticles underneath the ITO back contact could contribute to the effective absorption (Abs_{CIGSe}) of solar cells from an optical point of view.

To transfer the optical benefit to the electrical gain of solar cells, the next necessary step is to address the problem leading to the electrical failure of solar cells. Further, to achieve the

optimum gain of $\text{Abs}_{\text{CIGSe}}$ enhancement, the geometry of the Ag nanoparticles needs to be optimized, which covers the size, shape, density and periodicity.

Acknowledgements

The authors would like to thank R. Klenk for discussion, C. Ferber, M. Kirsch and J. Albert for technical support, C. Xiao for reading the paper. The authors acknowledge the funding from the Helmholtz-Association for Young Investigator groups within the Initiative and Networking fund (VH-NG-928) and G. Yin specially acknowledges the support of funding from China Scholarship Council.

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