Highly Transparent Singlet Fission Solar Cell with Multi-Stacked Thin Metal Contacts for Tandem Applications

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ABSTRACT:

Singlet fission solar cells combined with silicon photovoltaics allow the construction of parallel tandem solar cells, which benefit from better usage of high-energy photons. A key limiting factor for the performance of such a tandem configuration is the transparency of the singlet fission front cell. Here we show highly transparent singlet fission solar cells with a top contact of thin Ca:Ag blends. The optimized contact leads to 81% average solar cell transmittance in the near-infrared while maintaining more than half the short-circuit current density compared with an opaque device. We simulate the performance of the parallel tandem stack, and assess the improvements needed to fully realize the potential of singlet fission in this device configuration.

Charge carrier multiplication in solar cells allows the extraction of more than one charge carrier per absorbed photon.[1] This multiplication effect increases the theoretical power conversion efficiency (PCE) limit from 34 % to 45%.[2],[3] One of the most promising carrier multiplication mechanisms is a process known as singlet fission, where a spin-singlet exciton, generated by the absorption of a high-energy photon, spontaneously converts into two triplet excitons with half of the original singlet exciton energy. Small molecule organic semiconductors, such as pentacene, facilitate very high singlet exciton to triplet exciton conversion yields, approaching 200 %.[4],[5],[6] Thus, in configurations where these triplet excitons can be extracted, the photocurrent of the device can be increased.[7] One implementation of singlet fission based solar cells are parallel-connected tandem devices, where the singlet fission cell acts as the high-bandgap front cell in conjunction with a narrow-bandgap solar cell. Down-conversion via singlet fission facilitates voltage-matching between the high- and low-bandgap sub-cells. Recently, we demonstrated such a parallel connected tandem solar cell based on pentacene and crystalline silicon.[8] Such voltage-matched parallel connected tandem solar cells show more stable performance under a change of solar spectrum compared with the most common current matched series-connected tandem devices. However, parallel tandem solar cells did not achieve a high power conversion efficiency so far due to optical losses at the front solar cell. Ideally the front cell would absorb all photons above the bandgap of the singlet fission material, and be perfectly transparent at energies below that bandgap to maximize the number of harvested photons in the back cell. Therefore, the performance of the tandem stack is crucially determined by the transparency of the singlet fission front cell.

Here we investigate highly transparent, inverted pentacene solar cells with two transparent contacts, an ITO bottom contact and a thin metal layer stacked top contact. The semiconducting

singlet fission layer acts as a filter that absorbs high-energy photons, while being perfectly transparent to below bandgap radiation. Consequently, below bandgap transparency is critically limited by the near-infrared transparency of the electrodes in such devices. Therefore, we focus on the fabrication of a conductive and transparent top metal electrode. Indium tin oxide (ITO) is the most common material used for transparent contacts because of its remarkable optical transparency and electrical conductivity.[9] However, ITO deposition demands high power sputtering, which would decompose any underlying organic layer during the deposition process. Metal nanowire networks,[10],[11],[12] nano-meshes[13],[14] and carbon nano-structures based on carbon nanotubes[15],[16] and graphene[17] have shown sufficient optical transparency and electrical conductivity as a top electrode for semi-transparent organic solar cells. However, those nano-structured layers are assembled by wet chemical processing and thermal annealing to form a uniform and electrically conductive layer. A pre-deposited organic layer can be dissolved or degraded by solvents during those processes. Recently it was shown that multilayers of thin metals can lead to >90% of optical transparency with an electrical conductivity comparable to ITO.[18],[19],[20] This multilayer structure is easily prepared by a mild thermal evaporation process, which avoids the degradation of the underlying organic layers during the deposition. In spite of the advantages, multi-stacked metal electrodes are designed for hole-carrier collection, while conventional singlet fission based solar cells require electron extraction at the top electrode.

We introduce an inverted singlet fission based organic solar cell to incorporate the highly transparent multilayer electrode. The device architecture and energy band diagram of conventional and inverted singlet fission based organic solar cells are illustrated in **Figure 1**(a) and (b). For the conventional singlet fission cell we follow Congreve et al.,[6] using a

heterojunction between pentacene and C_{60} fullerene with poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) / poly(3-hexylthiophene-2,5-diyl) (P3HT) as the hole extraction layers. For the inverted cell we use molybdenum oxide (MoO_x) as the hole extraction (top) layer and bathocuproine (BCP) and zinc oxide (ZnO) as the bottom electron extraction layers. The current density - voltage (*J-V*) performance of opaque conventional and inverted solar cells are shown in **Figure 1**(c) and summarized in the inset. The conventional cell shows a short-circuit current density (J_{SC}) of 6.7 ± 0.3 mA/cm^2, which is higher than the J_{SC} of 5.1 ± 0.2 mA/cm^2 from the inverted cell. However, the inverted solar cell shows superior open-circuit voltage (V_{OC}) of 0.40 ± 0.01 V and fill factor (FF) of 56 ± 2 % in comparison to the conventional cell with a V_{OC} of 0.34 ± 0.01 V and a FF of 45 ± 1 %, resulting in 1.12 ± 0.1 % of average PCE.

The solar cell performance is significantly affected by the thickness of the pentacene layer. We optimized the thickness of pentacene in the inverted solar cell from 20 to 100 nm to find the ideal thickness, as shown in the J-V curves in **Figure 1**(d). J_{SC} increases with the thickness from 20 to 100 nm. The enhanced absorption of the thicker pentacene layer presumably leads to higher exciton generation rate while still allowing for most excitons to reach the pentacene/C_{60} interface. The V_{OC} of the solar cells is also increased when the pentacene thickness is increased, presumably because of voids forming in the thin pentacene films (see Supplementary Information S2). At higher thicknesses, the J_{SC} and V_{OC} drop due to recombination in the pentacene layer as the layer thickness exceeds the exciton diffusion length.[21],[22] Additionally, the thicker layers (≥80 nm) led to less reproducible results, presumably due to the rougher surface morphology of the thick layers (see Supplementary Information S2). In the following we will use solar cells with 60 nm of pentacene.

To optimize the optical transparency of the pentacene solar cell for the tandem device configuration, we introduced two different semi-transparent top electrodes composed of multi-stacked metals replacing the conventional opaque metal electrode. Firstly, we simply reduced the thickness of the Ag layer down to 10 nm. To achieve uniform Ag deposition we deposited 1 nm of aluminum (Al) underneath of Ag as a seed layer. The transmittance of the semi-transparent inverted pentacene solar cell with the thin Al/Ag contact is shown in **Figure 2**(a). Upon reducing the thickness of Ag from 70 to 10 nm the transmittance in the visible to near-infrared (NIR) region (400-1100 nm) increases from 5 % to 40 %. To reduce the reflection from the top electrode, we introduced 94 nm of molybdenum oxide (MoO_X) on top of the Al/Ag electrode as an anti-reflection (AR, $\lambda/4$ = 225 nm) coating considerably enhancing the transmittance of the device in the near-IR region (dashed lines in **Figure 2**(a)). **Figure 2**(b) shows the average transmittance of the full device in the visible region (400 to 800 nm, T_{vis}) and the region below the pentacene band gap which we call NIR region (700 to 1100 nm, T_{NIR}) as a function of Ag layer thickness. The transmittance continuously decreases with top contact thickness. With the AR coating both T_{NIR} and T_{vis} are enhanced by 5 - 15 %. The sheet resistance (R_S) of the contact has to be considered for efficient charge collection in a solar cell. Ag contacts of all thicknesses without AR coating present R_S below 6 Ω/□ except for the 10 nm Ag contact with much larger R_S of 127 Ω/□ (see **Figure 2**(c)), presumably because of the formation of unconnected Ag domains.[19]

The second semi-transparent multilayer metal contact we apply is a blend of calcium and silver (Ca:Ag). Schubert et al. showed that this combination of materials exhibits an unusually high optical transmittance, while maintaining good conductivity.[20],[23] We sequentially deposited 1 nm of Al and Ag seed layers on top of the solar cell structure to achieve a uniform Ca:Ag layer.

Ca and Ag were co-evaporated to form a blend that self-assembles as a Ag network surrounded by a Ca shell.[23] **Figure 2**(d) shows the transmittance of semi-transparent solar cells with varying Ca:Ag thicknesses. All devices with Ca:Ag electrodes show superior transmittance, almost 30 % higher over the entire wavelength range in comparison to devices with the same thickness of thin Ag electrode. As an AR coating we used 90 nm of tris(8-hydroxyquinolinato)aluminium (Alq_3) on top of the Ca:Ag electrode, following Schubert et al.[23] (dashed line in **Figure 2**(d)). The AR coating is designed to maximize T_{NIR} ($\lambda/4$ = 200 nm) and thus decreases the T_{vis}, while T_{NIR} is enhanced by around 10 % for all Ca:Ag thicknesses. The AR layer enhanced the transparency of the solar cell in the 700 to 1100 nm region even more significantly compared to the thin Ag electrode. The device with 10 nm of Ca:Ag electrode with AR coating achieved the maximum transmittance of 92 % at 820 nm. T_{vis} and T_{NIR} are decreasing with increasing Ca:Ag layer thickness, but with a much weaker thickness dependence compared to the thin Ag electrode (**Figure 2**(b) and (e)). The thickness of the Ca:Ag layer also determines the electrical performance, and in the following we optimize the solar cell for best overall performance. The MoO_x/Al/Ag/Ca:Ag structure shows higher R_S than the MoO_x/Al/thin Ag for all layer thickness (see **Figure 2**(f)). 40 nm of Ca:Ag contact allows 29 Ω/□ of R_S with 60 % of T_{NIR}, which is comparable to the 22 Ω/□ of our commercial ITO (125 nm), whereas it sharply increases to 140 Ω/□ at 10 nm of Ca:Ag contact, unfavorable for solar cell operation. We note that the resistance of our Ca:Ag contact is higher than reported previously for this electrode,[23] presumably because the surface roughness of the underlying active layer is larger (see Supplementary Information S2).

J-V characteristics of semi-transparent solar cells with thin Ag and Ca:Ag electrodes of varying thickness are shown in **Figure 3**(a) and (b), respectively. A semi-transparent solar cell

with a thin Ag (40 nm) electrode 0.90 % of PCE, shows a performance of only slightly less than the opaque solar cell (**Figure 3**(a), full solar cell details in Supplementary Information Table S3). The solar cell performance is almost identical with and without the AR coating (Supplementary Information S4). When the Ag thickness is reduced to 20 nm to yield higher transmission (T_{NIR} of 40 %) the solar cell performance is considerably reduced (PCE 0.45%), mostly due to a lower J_{SC} of 2.5 mA/cm^2. The Ca:Ag electrode allows for much higher T_{NIR} values for comparable solar cell efficiencies. The AR coated semi-transparent solar cell with a 50 nm thick Ca:Ag electrode presents 3.0 mA/cm^2 of J_{SC}, and a PCE to 0.64 % at 54 % of T_{NIR} (see **Figure 3**(b)). An extremely transparent solar cell with only 20 nm Ca:Ag layer shows 81 % of T_{NIR} while maintaining 2.3 mA/cm^2 of J_{SC}, yielding 0.50 % of PCE. We note that the reduction in J_{SC} in the transparent solar cell is mostly because of the absence of a reflective silver contact. The reflection of the silver contact allows designing the organic solar cell as a Fabry-Perot cavity, leading to much stronger absorption in the pentacene layer.[24] Transparent solar cells thus require materials with large triplet exciton diffusion length or selective reflectivity of the contact. **Figure 3**(c) summarizes the J_{SC} as a function of T_{NIR} with the two types of electrodes. The Ca:Ag layer yields nearly 20 % higher T_{NIR} for the same J_{SC} value than solar cells based on the Al/Ag contact. J_{SC} continuously decreases to half the value of the opaque device at 70 % of T_{NIR} owing to lower conductivity and reduced reflection off the back contact. V_{OC} and FF are almost identical for all contact thicknesses > 10 nm.

Semi-transparent singlet fission solar cells can be used as the front cell of a tandem device configuration in combination with a Si back cell. Singlet fission allows the connection of devices in a two-terminal parallel cell (**Figure 4**(a)), ensuring higher spectral stability compared to conventional, series connected tandem cells[8] while maintaining the two-terminal advantage

compared to four-terminal cells (**Figure 4**(b)). In the following we calculate the potential singlet fission/Si tandem solar cell efficiency based on the transmittance spectrum and the performance of the semi-transparent singlet fission front cells with different Ca:Ag thicknesses (see Supplementary Information S6 for details of the simulation).[25] **Figure 4**(c) shows the calculated J_{SC} of parallel tandems made from the singlet fission cells together with the record performing silicon solar cell.[26] A thick Ca:Ag contact enhances the J_{SC} of a semi-transparent front cell by lowering the R_S. However, more serious photon absorption and reflection at the thick electrode reduces the J_{SC} of the Si back cell. The J_{SC} of the parallel tandem solar cell is maximized at 32.6 mA/cm^2 for 15 nm of Ca:Ag contact thickness (**Figure 4**(d)). However, the tandem solar cells do not yield performances higher than the Si bottom cell alone, even with the highly transparent front cell obtaining >80 % of T_{NIR}. To achieve a tandem cell performance higher than the two individual sub-cells, the current of the front cell needs to be higher than the current loss from placing it in front of the silicon cell. In the final device configuration, the silicon and the singlet fission cell might share a common ITO contact. Some silicon cells already feature ITO contacts,[27] and the singlet fission cell could simply be placed on top. The AR coating of the Si cell would then need to be placed on top of the singlet fission cell. Thus, the absorption at the active layer of the singlet fission front cell needs to be maximized to benefit from carrier multiplication, while maintaining above unity quantum efficiency. In addition, the parallel tandem solar cell shows lower PCE in comparison to the four-terminal tandem because of the large voltage difference between the maximum power point (V_{mpp}) of the semi-transparent pentacene device and the Si solar cell (see Supplementary Information Figure S6). V_{mpp} matching is required in order to maximize the performance of parallel connected tandem solar cells. For this reason, singlet fission cells based on materials with a higher triplet energy (such as

tetracene), and thus larger voltage, would greatly benefit the tandem solar cell performance. However, in our hands, as well as in the literature, tetracene based singlet fission solar cells have shown lower quantum efficiency than pentacene cells,[28] despite the quantitative singlet fission in tetracene under optimized conditions.[29],[30],[31] This illustrates the requirement for optimizing device architecture, interfaces, and tetracene growth. Furthermore, the series resistance of the singlet fission solar cell is high in comparison with the Si solar cell, which degrades the FF of the parallel tandem (see Supplementary Information S2).

In summary, we have demonstrated semi-transparent pentacene solar cells introducing a highly transparent top contact, which could be used to sensitize a silicon solar cell or other low-bandgap cells in a parallel tandem configuration. We introduce an inverted singlet fission solar cell architecture, designed to allow for the transparent electrode, which achieves 1.21 % of PCE, higher than 1.03 % of conventional, non-inverted singlet fission solar cells. We characterize the inverted solar cells with thin Al/Ag and Al/Ag/Ca:Ag multi-stacked metals as a transparent electrode with various electrode thicknesses. Thin Ag shows low sheet resistance (below 6 Ω/□) while the Ca:Ag blend layer leads to a high optical transparency (e.g. 48 % of T_{NIR} with 28 Ω/□ sheet resistance). Furthermore, deposition of an anti-reflection layer on Ca:Ag enhanced the T_{NIR} of the solar cell. The *J-V* characteristics of semi-transparent solar cells were mainly determined by the thickness of the transparent top contact. In particular, the cell with a 20 nm Ca:Ag contact showed more than half the J_{SC} of the opaque solar cell, with 81 % T_{NIR}. Finally, we calculate the efficiency of a parallel tandem configuration consisting of our semi-transparent singlet fission front cells together with a Si back cell, absorbing high- and low-energy photons, respectively. The compromise between transmittance and performance of the front cell determined the J_{SC} of the tandem configuration, resulting in a maximum J_{SC} of 32.6 mA/cm^2. However, reaching a

higher tandem performance than the Si cell alone is still challenging using the current semi-transparent singlet fission front cells. Enhancing the absorption in the active layer is critical to maximize carrier multiplication, especially for singlet fission materials with high triplet energy to improve the V_{OC} of the singlet fission cell and the overall tandem performance.

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Supporting Information Available: We provide details about fabrication and characterization of the semi-transparent singlet fission solar cells and the model used to simulate the performance of singlet fission/Si tandem solar cells.

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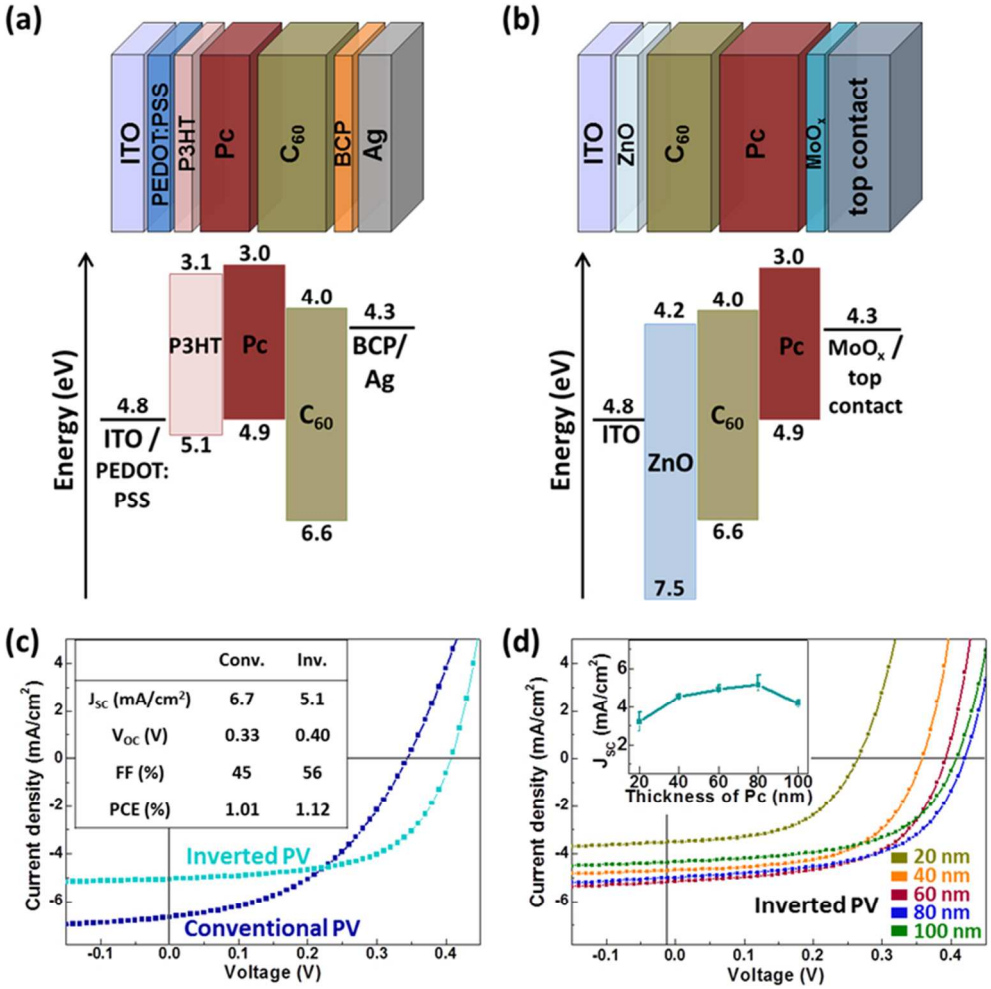

Illustration of the solar cell architecture and energy band diagram of (a) the conventional pentacene solar cell based on reference[6] and (b) the inverted solar cell with different top contact structures such as the opaque silver (100nm thick), transparent thin silver and transparent calcium:silver blended layer. (c) Current density-voltage (*J-V*) characteristics of conventional and inverted solar cells (d) *J-V* characteristics and short-circuit current density (J_{SC}, inset) of inverted solar cells with 20 to 100 nm of pentacene layer thickness.

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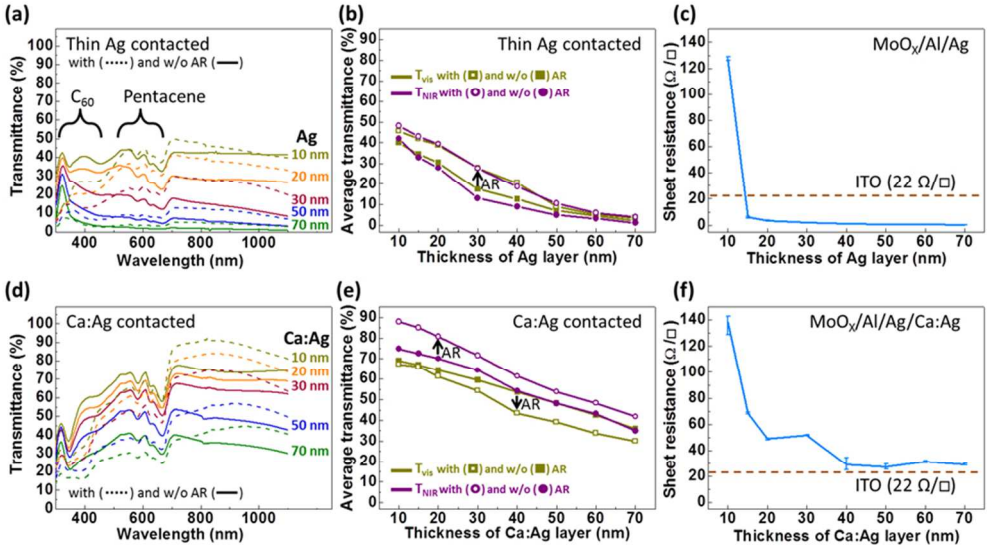

Optical transmittance of solar cell with different thickness of (a) thin Ag contact and (d) Ca:Ag contact. Brackets in (a) indicate mostly C_{60} and Pentacene absorption. Calculated average transmittance of solar cell in visible (400 to 800 nm, T_{vis}) and near infrared (700 to 1100 nm, T_{NIR}) region with (b) thin Ag contact and (e) Ca:Ag contact with and without anti-reflection (AR) coating. Sheet resistance as a function of layer thickness of (c) Ag and (f) Ca:Ag without AR coating. The dashed lines shows the sheet resistance of our commercial ITO (125 nm).

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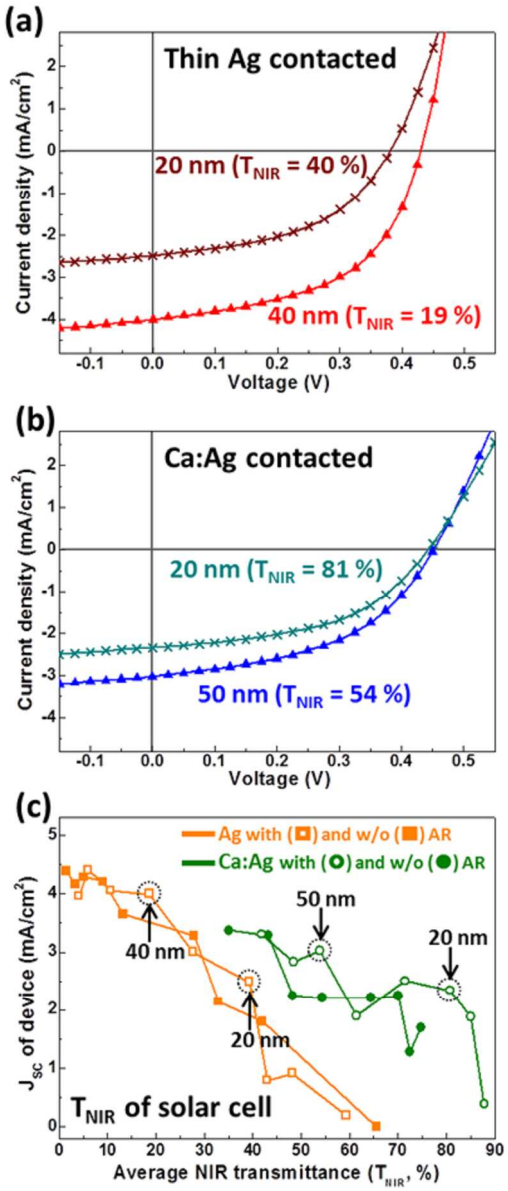

J-V characteristics of semi-transparent solar cells with (a) thin Ag contact and (b) Ca:Ag contact. (c) J_{SC} of semi-transparent solar cells with the two types of top contact as a function of T_{NIR} for solar cells with and without AR coating.

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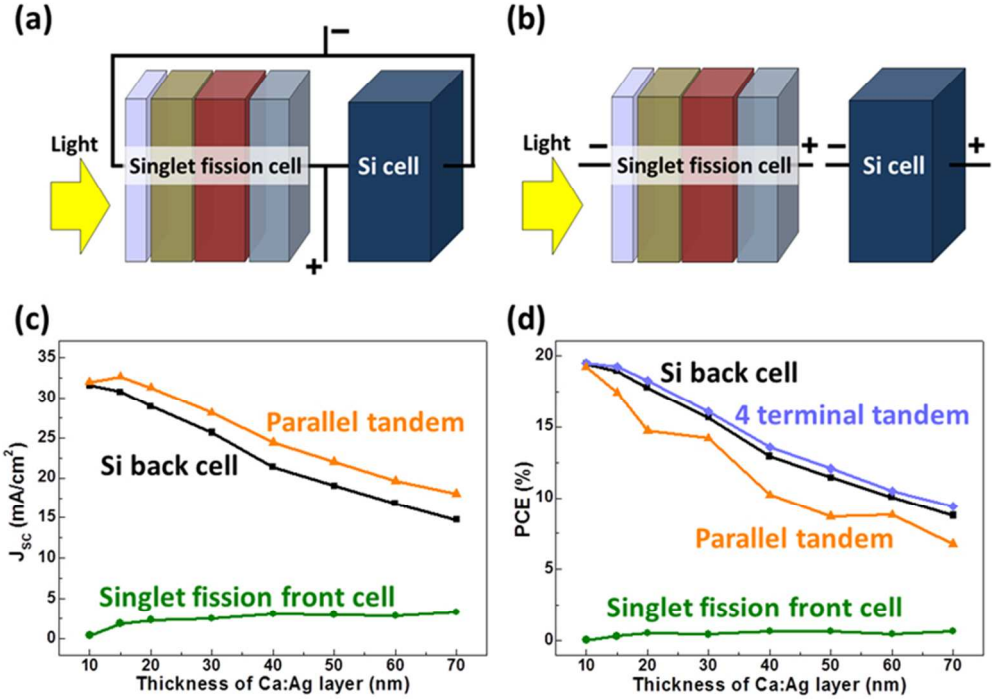

Illustration of tandem configuration based on semi-transparent singlet fission front and silicon back cells. Sub-cells are electrically connected in (a) parallel and (b) four-terminal. (c) Calculated J_{SC} and (d) PCE of singlet fission/Si tandem solar cells from our transparent pentacene cells with different Ca:Ag layer thicknesses.

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