

钙钛矿/硅叠层太阳能电池：见解与展望

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摘要

有机-无机杂化钙钛矿具有可调带隙、高光吸收系数和较高的功率转换效率等优点, 在硅基叠层太阳能电池中得到了广泛的应用。然而, 钙钛矿/硅叠层太阳能电池的最大功率转换效率仍低于理论极限。本文介绍了钙钛矿/硅叠层太阳能电池的工作原理、对钙钛矿/晶硅叠层电池的类别、影响其性能的主要因素进行了归纳综述。此外, 还强调了, 对于双面性的叠层电池结构, 4-T叠层可能比2-T叠层更具吸引力。

关键词

叠层太阳能电池, 双面叠层太阳能电池, 寄生吸收, 反射损耗, 电流匹配

Perovskite/Silicon Tandem Solar Cells: Insights and Outlook

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Abstract

Organic-inorganic hybrid perovskites have been extensively used in silicon-based tandem solar cells due to their adjustable bandgap, high light absorption coefficient, and enhanced power conversion efficiency. However, the maximum power conversion efficiency of perovskite/silicon tandem solar cells remains below the theoretical limit. This study introduces the working principle of perovskite/silicon tandem solar cells and provides a comprehensive review of the types of perovskite/crystalline silicon tandem cells and the primary factors affecting their performance. Additionally, it emphasizes that for bifacial tandem cell structures, 4-T tandems may be more attractive than 2-T tandems.

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Keywords

Tandem Solar Cells, Bifacial Tandem Solar Cells, Parasitic Absorption, Reflection Loss, Current Matching

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1. 引言

用于大规模发电的太阳能电池被认为是取代传统化石能源和提供清洁电力的重要技术之一[1]。晶体硅太阳能电池因其高效率、优异的稳定性以及成熟的制造工艺，当前在光伏市场中占据主导地位。然而，随着其功率转换效率逐渐逼近理论效率极限的 33.5% [2]，进一步提高效率将变得愈发困难[2] [3]。

有机-无机卤化物钙钛矿太阳能电池因其高光吸收系数、长载流子扩散长度和可调带隙(1.17~2.8 eV)而受到广泛关注[4] [5]。凭借这些优异的光电特性，这类新型半导体推动了新一代太阳能电池的发展。单结钙钛矿太阳能电池的转换效率在十几年间取得了显著提升，从最初的 3.8%迅速跃升至 26.1%，如图 1 所示[6]。然而，在单结太阳能电池中，仅有一部分太阳能能够被吸收并转化为电能。被吸收的光子中，尤其是高能量光子的大部分能量会转化为热量，导致损耗增加[7]。因此，提高从太阳光谱中提取光子的效率至关重要。

叠层电池通过组合不同带隙的电池，实现了更高效的光吸收和转换，从而有效提高了功率转换效率，减少能量损耗[8] [9]。钙钛矿太阳能电池可以与中带隙或窄带隙的底层电池匹配，例如硅电池、窄带隙钙钛矿电池、有机太阳能电池、铜锌锡硫化物(CZTS)以及铜铟镓硒(CIGS)太阳能电池[10]-[12]。在这些组合中，钙钛矿/硅叠层太阳能电池技术的发展速度最快，其效率已从最初的 13.7%提升至 34.6% (图 1) [6]。尽管在效率方面取得了这些重大进步，但要达到这种组合的实际极限(41%)还有很长的路要走[13]。

尽管已有综述探讨了钙钛矿/硅叠层太阳能电池的发展历史及其稳定性[1] [6] [9] [14]-[16]，但目前对于该叠层电池的分类及其效率的关键制约因素仍缺乏系统性分析。本文介绍了钙钛矿/硅叠层太阳能电池的工作原理、对钙钛矿/晶硅叠层电池的类别、影响其性能的主要因素进行了归纳综述。此外，还强调了，对于双面性的叠层电池结构，四端(4-T)叠层可能比两端(2-T)叠层更具吸引力。

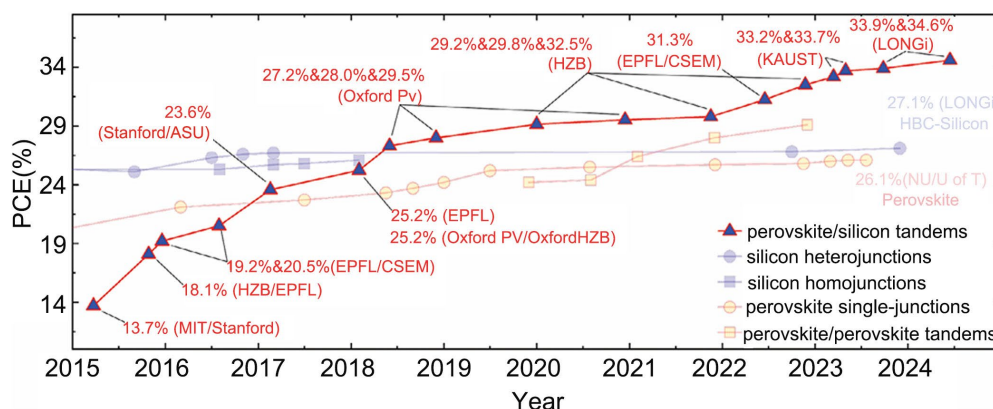


Figure 1. The efficiency of perovskite/silicon tandem solar cells has evolved significantly over the past eight years [6]

图 1. 8 年来钙钛矿/硅叠层太阳能电池效率的演变[6]

2. 钙钛矿/晶硅叠层太阳能电池的工作原理

叠层钙钛矿/硅太阳能电池中, 顶电池由宽带隙的钙钛矿太阳能电池构成, 主要吸收波长小于 800 nm 的高能量光子, 而底电池则由窄带隙的硅电池构成, 专门吸收波长小于 1100 nm 的低能量光子, 实现了对太阳光谱的分段利用, 这避免了单结器件中高能光子的热化损失, 并提高了电池的太阳能利用率和功率转换效率(图 2(a)~(b)) [17] [18]。

在 2-T 叠层中的两个子电池是光学和电连接的, 其中顶底子电池共用中间电极。所以通过两个子电池的电流密度(J)必须相同, 即“电流匹配”。2-T 叠层的功率密度(P_{2T})为[17]

$$P_{2T} = (V_1 + V_2)J. \quad (0.1)$$

其中 V 是子电池的电压。下标 1 和 2 分别表示顶和底电池。

2-T 叠层中“电流匹配”的要求为实现高效的 2-T 叠层带来了许多挑战, 包括两个子电池的带隙匹配和光子管理等。

在 4-T 叠层中, 两个子电池是独立的完整的太阳能电池。由于 4-T 叠层电连接不是串联的, 因此它们不受电流匹配的限制, 对带隙组合适应不太敏感。4-T 叠层的功率密度(P_{4T})为[19]

$$P_{4T} = P_1 + P_2 = V_1 J_1 \quad (0.2)$$

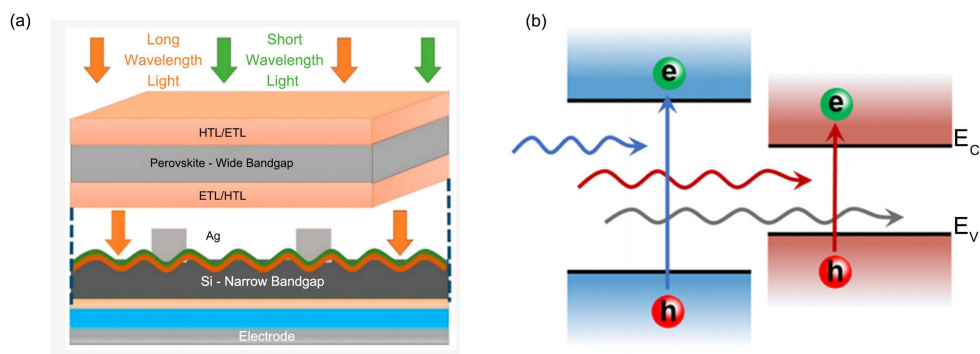


Figure 2. (a) Working principle of the tandem perovskite/silicon solar cell [18]; (b) Schematic diagram of light absorption for the two cells [17]

图 2. (a) 叠层钙钛矿/硅太阳能电池的工作原理[18]; (b) 两个电池的光吸收示意图[17]

3. 钙钛矿/晶硅叠层太阳能电池分类

2-T 结构和 4-T 结构是钙钛矿/晶硅叠层太阳能电池的两种主要典型结构。通过在这些结构中引入反射层, 形成双面叠层, 可以提高载流子的生成率, 从而增加能量输出。

3.1. 2-T 和 4-T 钙钛矿/晶硅叠层太阳能电池

两个子电池堆叠构成叠层太阳能电池, 利用不同波段的光谱以突破单结的 Shockley-Queisser 极限[20]。其主要配置分为 2-T 和 4-T 两种。在 2-T 结构中, 顶子电池直接生长在底子电池上, 这要求严格的制造兼容性和电流匹配, 同时较少的透明电极有助于减少反射和寄生吸收[21]。相比之下, 4-T 电池是通过将一个半透明的宽带隙顶电池机械叠加到窄带隙底电池上来构建的[22]。在此配置中, 顶电池和底电池在电气上相互隔离, 允许独立优化并分别提取功率输出。这样的设计有效提升了光吸收和能量转换效率。

在 2-T 叠层钙钛矿/硅太阳能电池中, 两个子电池串联连接(如图 3(a)) [23], 其中钙钛矿顶电池通过重组层结合底电池上。由于电流匹配的要求, 顶电池的带隙被限制在 1.7 eV~1.8 eV [24]。同时, 为了避免

对底电池造成损害,顶电池的制造也比较复杂。由于该叠层设计仅有两个端子,便于在光伏模块和系统中集成,并降低了制造成本。然而,电流匹配仍然是该设计中的主要问题,因为两个子电池是串联连接的。Qiu 等人[25]研究了四种不同钙钛矿材料的 2-T 叠层电池。他们通过调整钙钛矿吸收层的带隙和厚度,实现了两个子电池之间的电流匹配。其中,使用带隙为 1.69 eV 的钙钛矿的叠层电池达到了 22.2% 的功率转换效率。Xu 等人[11]使用三卤化物合金(Cl、Br、I)策略获得了带隙为 1.67 eV 的钙钛矿,并有效抑制了光诱导的相分离。掺入氯的钙钛矿显示出两倍的载流子寿命和电荷载流子迁移率,从而使 1 cm²面积的 2-T 叠层太阳能电池达到了 27% 的效率。Kim 等人[26]则通过阴离子工程开发了一种高效且稳定的叠层太阳能电池。他们在钙钛矿前驱体中添加了基于苯乙胺的添加剂 PEA (I_{0.25}SCN_{0.75}),其中 SCN⁻促进了晶粒的生长。同时,使用 PEA (I_{0.25}SCN_{0.75})添加剂在表面和晶界形成了 2D 铅碘化物框架,从而实现了更好的钝化。该叠层太阳能电池的功率转换效率达到了 26.7%。目前,通过采用自组装单分子层(SAM)作为空穴选择性层[27],以及先进的光学优化,叠层太阳能电池的效率进一步提升,达到 32.5%。最新的记录效率为 34.6% [28]。

在 4-T 叠层钙钛矿/硅太阳能电池中与 2-T 结构相似(如图 3(b)) [23],两个子电池是垂直堆叠的,但它们在光学上耦合而电气上独立。每个子电池都有自己的独立两个端子,这使得它们可以单独操作和优化,从而减轻了顶电池带隙选择的限制,降低了系统对光谱变化的敏感性[29]。因此,4-T 叠层电池能够在 1.6 至 2 eV 的广泛带隙范围内实现高效率,当与 c-Si 底电池组合时,顶电池最佳带隙为 1.81 eV [30]。该设计不需要电流匹配和子电池之间的界面或重组层,但需要多个透明电极,这可能导致更多的寄生吸收。此外,因设计中需要更多电缆和最大功率点跟踪器,整体制造成本可能会增加[31]。Loper 等人[32]于 2014 年首次报道了 4-T 叠层钙钛矿/硅太阳能电池。他们采用 MAPbI₃ 作为顶电池, c-Si 异质结作为底电池。在透明的 MoOx/ITO 电极下,该 4-T 叠层电池的功率转换效率达到了 13.4%。Chen 等人[33]通过在 1 nm 厚的铜层上沉积 7 nm 金,开发了一种带金属电极的半透明钙钛矿。这种半透明钙钛矿的效率为 16.5%,当叠加在硅异质结电池上时,4-T 叠层器件的总效率达到了 23.0%。然而制备具有合适带隙的高质量钙钛矿对于 4-T 叠层器件也非常重要。Duong 等人[34]通过在 CsFAMA 体系中引入铷(Rb),开发了带隙为 1.73 eV 的钙钛矿。这种电池几乎没有迟滞现象,并且光稳定性得到了提升。在 720~1100 nm 波段,半透明电池的功率转换效率为 16%。在 4-T 叠层器件中,该电池的效率达到了 26.4%。Sargent 等人[35]采用增强溶剂萃取法沉积了光滑且较厚的钙钛矿薄膜,从而提高了光程长度。该钙钛矿太阳能电池的功率转换效率为 19.2%,在近红外波段的平均透过率为 85%。将这种电池与硅电池机械堆叠后,4-T 叠层器件的效率达到了 28.2%。

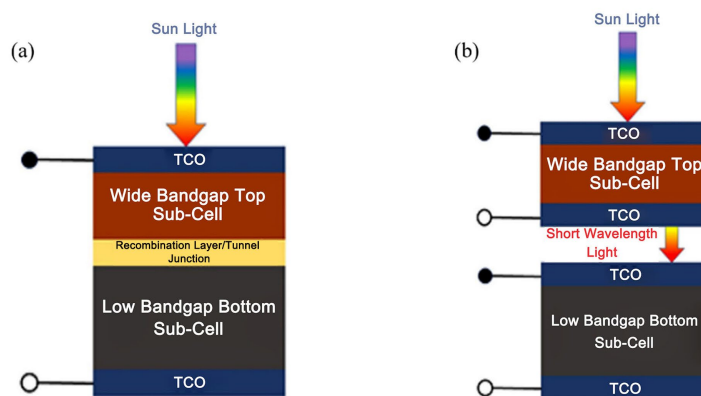


Figure 3. Device structure of tandem perovskite/silicon solar cells [23]: (a) 2-T structure; (b) 4-T structure

图 3. 叠层钙钛矿/硅太阳能电池器件结构[23]: (a) 2-T 结构; (b) 4-T 结构

3.2. 双面钙钛矿/硅叠层太阳能电池

双面光伏技术能够进一步提升 c-Si 光伏组件的能量产量, 通过使用硅异质结技术可以轻松实现这一点, 只需将不透明的后金属接触层替换为网格化金属化结构[36]。由于电池的背面是透明的, 它可以收集来自周围环境的反射和散射光(即地表反射光或反照率), 从而增加发电量[37]。如图 4(a) [38]。对于优化的单结器件, 产生的电流会随着反照率的增加而线性增长。在硅太阳能电池技术上, 通过利用来自环境的反射光, 将功率输出提高 25%到 30%, 预计到 2030 年将占据光伏市场的 85%份额[39]。计算结果表明, 钙钛矿/硅叠层电池同样可以从双面性中受益[37] [40] [41]。实验研究报告显示, 一种双面 4-T 钙钛矿/硅叠层太阳能电池在实际反射率为 20%的情况下实现了 30.5%的光电转换效率[42]。同样, Bastiani 等人[38]证明, 在相同的反射率条件下, 一种双面 2-T 叠层达到了 25.5%的效率, 相比于单面电池, 其发电量显著增加了 19%。开发有效的双面叠层技术的关键在于选择合适的器件材料和结构。2-T 和 4-T 结构(图 4(b)~(c)) [42]在实现高效双面叠层太阳能电池方面各有优缺点。无论是哪种叠层结构, 关键的设计步骤都是确定最合适的子电池带隙。在 2-T 结构中, 双面叠层电池需要针对这一目的对钙钛矿的带隙进行重新设计[40]。由于顶部和底部电池的带隙分别较大和较小, 反照率的增加主要会提高底部电池产生的电流, 这种情况可能导致电流不匹配[43], 从而降低功率转换效率, 并可能对系统级性能产生显著影响[40]。因此, 在进行传统的电流匹配优化时, 应考虑通过调整钙钛矿顶部电池的厚度和带隙来适应反照率的影响[44] [45]。然而, 在长时间的光照射下, 这种调整可能导致相分离, 从而导致器件退化[46]。最近有研究探索了 4-T 叠层电池, 与 2-T 叠层相比, 4-T 叠层具有以下四个优势, 使其在双面叠层应用中更具适应性: 对反照率的敏感度较低、理论效率(EYmpp)较高、对带隙变化的容忍度较高, 以及更低的模块更换成本。因此, 4-T 叠层在双面叠层应用中比 2-T 叠层更为适合[29]。

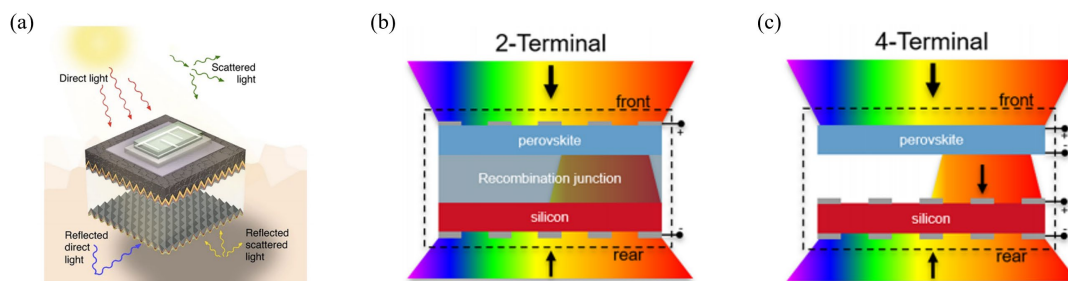


Figure 4. (a) Schematic diagram of light absorption for the layered structure with reflectivity characteristics [38]; (b) Bifacial 2-T tandem device [42]; (c) Bifacial 4-T tandem device [42]

图 4. (a) 具有反照率特征的叠层结构的光吸收示意图[38]; (b) 双面 2-T 叠层器件[42]; (c) 双面 4-T 叠层器件[42]

4. 影响高效钙钛矿/晶硅叠层太阳能电池光电转换性能的关键因素

4.1. 寄生吸收损耗

寄生吸收是指光子在非活性层中被吸收, 无法有效利用, 从而导致光损失并降低电池的功率转换效率。在 2-T 钙钛矿/硅叠层太阳能电池中, 寄生吸收损失主要发生在两个子电池的中间连接层和钙钛矿电池的空穴传输层中。透明导电氧化物(TCOs)通常用作复合层, 以提高太阳能电池的功率转换效率。在众多 TCO 材料中, 铟锡氧化物(ITO)因其优良的光伏特性而被广泛应用[47]。然而, ITO 在近红外波长范围(800~1200 nm) [47] [48]内存在载流子吸收, 这会减少传递到底部电池的光子数量, 导致近 5%的寄生吸收损失。为了解决由 ITO 引起的寄生吸收问题, 许多研究人员开始使用 ZTO [49] (见图 5(a))和 nc-Si:H [50] (见图 5(b))等材料替代 ITO。这些替代材料虽然载流子浓度较低, 使得载流子在薄膜中能够快速移动。此

外使用具有低寄生吸收的电子传输层(ETL)作为 p-i-n 结构钙钛矿太阳能电池的窗口层,可以避免寄生吸收效应[51]。Bush 等人[52]提出的使用 SnO_2 作为 ETL, 具有寄生吸收最小和高效电子提取速率的优点。同样, Ballif 等人[53]的最新研究展示了一种高效器件, 使用 C60 作为 ETL, 其电子迁移率高, 获得了 31.25% 的认证功率转换效率。

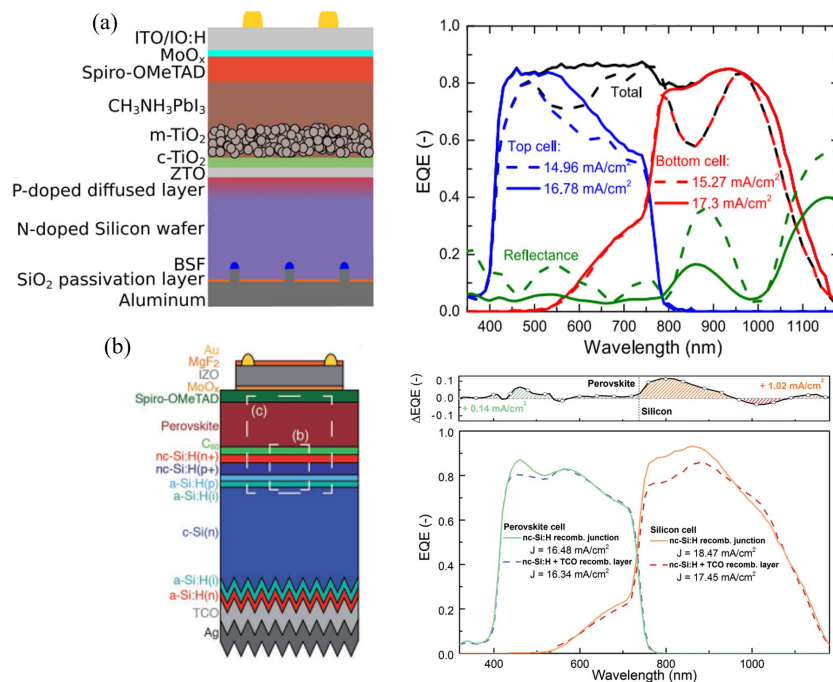


Figure 5. Schematic diagram of perovskite/silicon tandem solar cell structure: (a) ZTO as the interlayer (left), reducing parasitic absorption (right) [49]; (b) nc-Si:H as the interlayer (left), reducing parasitic absorption (right) [50]

图 5. 钙钛矿/晶硅叠层太阳能电池结构示意图: (a) ZTO 为连接层(左), 减少寄生吸收(右) [49]; (b) nc-Si:H 为连接层(左), 减少寄生吸收(右) [50]

4.2. 反射损失

反射损失指的是当光子照射到设备表面时, 由于设备光滑表面的反射或不同材料之间的折射率差异, 部分光子被反射或损失, 导致设备的功率转换效率降低。在钙钛矿/硅叠层太阳能电池中, 光学损失主要发生在平坦的前电极表面、硅电池表面以及复合层中的透明导电氧化物(TCO), 这些损失占据了光学损失的很大一部分。为了提高叠层的光吸收效率, 可以采取以下三种措施: 先调整 TCO 的折射率, 以减少反射损失; 由于钙钛矿(约为 2.4)与硅(约为 3.7)之间的折射率差异, 近红外光的反射增加, 从而减少到达硅基底电池的光的光学吸收, 导致硅基底电池的光电流下降。具有适中折射率和低寄生吸收的中间层可以减少光的反射, 增强光进入底电池的光学耦合[54]。nc-SiO_x 作为复合层材料, 在 800 nm 时的折射率为 2.6, 位于钙钛矿和晶体硅之间, 能够减少近红外光的反射。Mazzarella 等人[55]将 nc-SiO_x 用作钙钛矿/硅异质结叠层电池的复合层, 减少了底电池对近红外光的反射, 提高了硅基底电池的短路电流密度。使用 nc-SiO_x 的最佳叠层器件在 1 cm² 面积下实现了 38.7 mA/cm² 的电流和 25.2% 的功率转换效率, 如图 6(a)~(b) 所示。其次, 在叠层的前电极表面设计抗反射涂层(ARC), 增强其抗反射能力, 从而降低光的逃逸; 常用的 ARC 材料包括高透明度和低折射率的材料, 如光管理(LM)薄膜[56]、氟化锂(LiF) [57]和氟化镁(MgF₂) [58]。将 LM 薄膜应用于太阳能电池的光滑表面, 可以有效降低反射损失。同时, 并通过在 LM 薄膜与空气之间的界面上对向上传播的光进行反向散射, 增强了光的束缚, 从而提高了太阳能电池的光吸收效率

[59]。Albrecht 等人[56]表明, 引入 LM 薄膜后, 顶部和底部电池的短路电流密度都有所增加, 分别为 2 mA/cm^2 和 0.92 mA/cm^2 。当 LiF 和 MgF_2 等无机材料常被用作 ARC 时, MgF_2 和 LiF 具有低吸收率且易于制成更薄的层, 从而对入射光表现出更好的抗反射效果[60]。Albrecht 等人[61]通过在叠层电池顶部添加热蒸发的 LiF 抗反射层, 减少了叠层电池在空气/ITO 界面的反射损失, 导致硅电池的光生电流增加了 1.5 mA/cm^2 。Bett 等人[62]利用热蒸发方法在叠层电池前面施加 90 nm 的 MgF_2 作为 ARC, 以减少叠层电池前侧的反射。第三, 采用纹理化的硅基底, 减少表面的光反射损失, 最大限度地增加光子进入硅基底的数目。最近的许多研究表明, 叠层电池中的硅电池表面的纹理化可以显著抑制反射损失, 并提高器件的短路电流密度和光电转换效率[58] [63]。例如, Kanda 等人[64]研究了两种类型的叠层电池(一个硅基底为平坦的, 一个硅基底为纹理化的), 并比较了它们的性能。他们发现, 与平坦的硅表面相比, 纹理化的硅表面在 $750\sim 1050 \text{ nm}$ 波长范围内更有效地减少了反射, 改善了器件的电流(见图 6(c)~(d))。

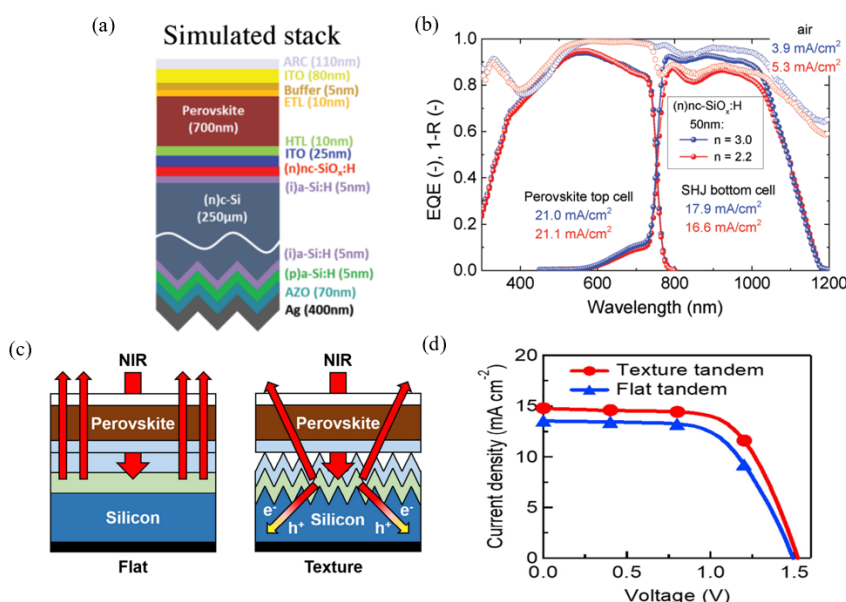


Figure 6. (a) Tandem cell with nc-SiO_x as the composite layer [55]; (b) Corresponding reduction in reflectance absorption [55]; (c) Schematic diagram of tandem structures with flat and textured silicon cells [64]; (d) Current of tandem cells with flat and textured silicon cells [64]

图 6. (a) 以 nc-SiO_x:H 为复合层的叠层电池[55]; (b) 相应的反射吸收减少[55]; (c) 具有平面和纹理硅电池的叠层结构示意图[64]; (d) 平面和纹理硅电池的叠层电池的电流[64]

4.3. 电流匹配

两个子电池之间良好的电流匹配对于实现叠层电池的最大效率至关重要。2-T 叠层电池总电流总受到电流较低的子电池的限制, 从而降低叠层电池的性能[65] [66]。Kohnen 等人[66]通过光学优化和精细调整钙钛矿吸收层、顶部电极和中间层的厚度, 改善了钙钛矿/硅叠层电池的性能, 以确保两个子电池之间的电流密度匹配。除了调整厚度之外, 使用透明电极作为前表面电极[67]、设计连接层[68]、优化硅基底电池的表面形态[63] [69]以及其他策略都可以实现更好的电流匹配[56]。然而, 最近有研究指出 2-T 叠层的最大效率与最佳电流匹配点发生偏移[65] [70] [71] (见图 7)。这种轻微的偏差通常归因于短路电流的降低和填充因子的补偿效应, 即在接近电流匹配条件时, 短路电流达到最大值而填充因子达到最小值。为了再现和解释这一现象, 采用了等效电路模型, 通过结合每个子电池的串联电阻、并联电阻和缺陷电阻等关键参数, 可以有效调节叠层太阳能电池的性能[72] [73]。例如, 根据 Kohnen 等人[66]的模拟计算,

电流不匹配可能伴随着叠层电池填充因子的增加。因此，均匀的光电流性能对于高效钙钛矿/硅叠层电池的光学设计至关重要。

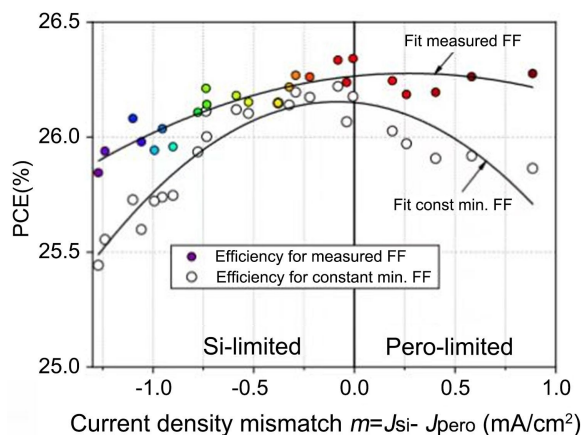


Figure 7. Impact of current offset on tandem efficiency [65]

图 7. 电流发生偏移对叠层效率的影响[65]

5. 展望

两个子电池堆叠构成叠层太阳能电池，通过利用不同波段的光谱来突破单结的 Shockley-Queisser 极限。然而，目前钙钛矿/硅叠层太阳能电池的最大功率转换效率仍低于理论极限。在现有技术基础上，提高叠层电池效率、减少寄生吸收损失、优化光子管理以及选择合适的电池架构，成为突破其理论效率的关键问题。叠层结构的引入不仅提高了光谱的利用率，还增加了光吸收的范围。此外，叠层设计中的双面性使得电池可以同时从正面和背面接收光照，这进一步提升了光子捕获效率和整体能量输出。因此，如何优化双面设计以最大限度地利用光谱，以及改善光子管理策略，成为进一步提高钙钛矿/硅叠层太阳能电池效率的重要研究方向。

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