

occur when deliberately using specific non-stoichiometric growth conditions.

Until now, it had been generally thought that Ruddlesden–Popper phases grow in the same layer-by-layer order that their constituent monolayers are deposited. These latest results undermine this assumption, with dramatic consequences for the controlled production of high-quality thin films and their resulting properties. Understanding of the dynamic layer-rearrangement is a prerequisite for engineering atomically sharp interfaces at the desired location within the heterostructure. Based on these results, novel growth routes can now be envisaged, enabling the study of new interfacial phases containing any desired member of the Ruddlesden–Popper series, which can, in turn, lead to improved performance and new functionalities. Such growth routes are not only limited to layered oxides such as Ruddlesden–Popper phases, but can also be used to explore the

structure and properties of oxide interfaces more generally.

To study the fundamental physics of the dynamic rearrangement in detail, a strong degree of collaboration between experimental and theoretical solid-state scientists was necessary. Both teams used an oxide molecular beam epitaxy system with *in situ* growth-monitoring capabilities for the fabrication of Ruddlesden–Popper phases. Their crystalline properties, as well as the atomic stacking, were studied using state-of-the-art synchrotron X-ray scattering and high-resolution scanning transmission electron microscopy. Moreover, to support the experimental evidence for the rearrangement of the surface layer, density functional theory calculations were performed. It is by combining various experimental approaches with theory that provides the insight and mechanistic understanding that no individual research group or institution can hope to achieve alone.

Taken as a whole, these two studies clearly underline the role of thermodynamic factors during the atomic-layer-by-layer growth of complex materials, and provide a clear route for expanding the base of materials that can be prepared by design, with precise interface control. The range of quantum phenomena accessible in oxide interfaces is already enormous¹. With an even richer variety of oxide interfaces to explore, this will continue to be fertile ground for fundamental studies in the future. □

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PEROVSKITE SOLAR CELLS

Continuing to soar

The dream of printing highly efficient solar cells is closer than ever to being realized. Solvent engineering has enabled the deposition of uniform perovskite semiconductor films that yield greater than 15% power-conversion efficiency.

Michael D. McGehee

The emergence over the past four years of solar cells made by solution casting thin films of light-absorbing methylammonium lead halide perovskite semiconductors has captured the attention of materials scientists and researchers in renewable energy^{1–3}. The 15% efficiency values reported nearly a year ago ignited an explosion of research in this field (Fig. 1). Unsurprisingly, researchers quickly identified challenges that needed to be addressed for these materials to be technologically successful. Writing in *Nature Materials*, Nam Joong Jeon and colleagues⁴ now report a deposition approach that, by significantly improving the morphology control of the perovskite layers, provides a feasible strategy to overcome some of these problems and raises expectations for these semiconductors.

Casting uniform thin films of perovskites using the originally reported procedures is not trivial. These perovskites, unlike other commonly cast materials like polymers, are made from ionic solutions that exhibit low viscosities and lead to rapid formation of perovskite crystals. In many cases, as the crystals grow the material dewets from the surface — meaning that it is pulled

away from the regions surrounding the crystals — and openings in the film are formed. These openings cause the electrical contacts — which, in a common photovoltaic architecture, are conducting films deposited on top and on the bottom of the light-absorbing layer — to touch each other, forming an electrical short that lowers the

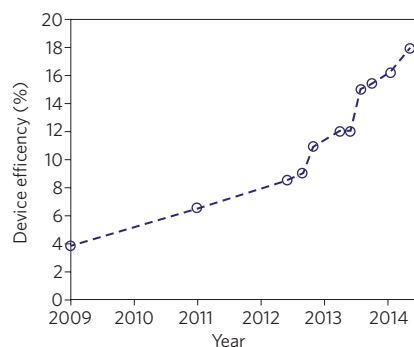


Figure 1 | The world record power-conversion efficiency of perovskite solar cells over time. Earlier data points were reported in the literature, but not validated. More recent data has all been validated by Newport or the National Renewable Energy Laboratory¹¹.

voltage of the cell. The variability in voltage caused by poor control of the formation of these openings has raised concerns about the possibility to make large solar panels based on these materials. Jeon and co-workers have resolved this dewetting problem by casting an intermediate thin film containing an extra-coordination solvent⁴. This precursor film fully wets the surface and can slowly be converted into the perovskite film by thermal annealing. The resulting films are remarkably uniform and have enabled the fabrication of solar cells with an average power-conversion efficiency of 15.5% and a relatively small standard deviation around 1%.

The researchers also address another problem made apparent during the past year: the hysteretic behaviour observed in the current–voltage curves of perovskite solar cells^{5,6}. For reasons that are not yet understood, the current often depends on the bias voltage applied to the cell seconds before the measurement is taken. Consequently, a current–voltage curve taken with increasing voltage (forward scan) tends to exhibit lower currents at each voltage than a curve taken with decreasing voltage (reverse scan; Fig. 2a). It is quite possible

that some of the efficiencies reported in the literature have been artificially enhanced by hysteretic effects, and that these solar cells would not demonstrate the reported efficiency if they were held at a steady voltage for an extended period of time. Jeon and colleagues investigated the behaviour of the layers deposited with their method by reporting current–voltage curves in the forward and reverse scanning directions. They demonstrated that hysteresis can be avoided by infiltrating the perovskite into a mesostructured scaffold of titania (Fig. 2b). However, it is not clear why the mesostructured titania reduces the hysteresis with respect to the perovskite films deposited on flat substrates; further investigation of the role of the perovskite/titania interface as well as of the dynamics of the charge carriers in these architectures is needed to fully understand this anomalous behaviour.

Although the results reported by Jeon and colleagues provide encouraging solutions to deposit uniform perovskite films and construct hysteresis-free solar cells, other issues remain to be addressed before perovskite solar cells can be turned into a game-changing technology. Through a combination of optimization strategies aimed at fine-tuning the perovskite chemical composition, improving perovskite film quality, tuning the energy levels of the charge-extraction materials used in the cell, and harvesting a broader part of the solar spectrum by using stacked cells, the power-conversion efficiency has the potential to soar past 20%, thus making perovskites competitive with the silicon solar cells that currently dominate the market. Bearing this in mind, efficiency limitations are unlikely to be the problem that slows the progress of perovskite solar cells towards commercial viability. Long-term stability, however, still presents a potential barrier for the success of perovskite semiconductors. Metal–halogen bonds tend to be weak and can sometimes

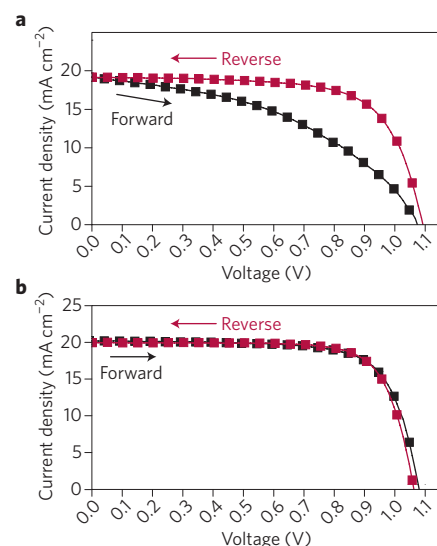


Figure 2 | The hysteresis problem in perovskite solar cells. **a**, Current–voltage curves with forward and reverse scans for a planar solar cell showing hysteresis. **b**, Current–voltage curves with forward and reverse scans for a mesostructured solar cell showing negligible hysteresis. Figure reprinted from ref. 4, Nature Publishing Group.

be broken when a photon is absorbed. This property is helpful for making photographic film, but not desirable for solar cells. We already know that these perovskites are not stable in the presence of humidity as they are hygroscopic, but we do not know how long they can last when properly encapsulated. Close behind stability are toxicity concerns; the general populace is unlikely to embrace a water-soluble lead compound for use in solar panels. Because of these concerns, the coming year will likely bring significant research on perovskite stability and the development of non-toxic perovskites — some of which has already begun as demonstrated by reports of successful substitution of tin in place of lead^{7–9}.

From a scientific perspective, it is fascinating to ponder why it is so easy to make highly efficient solar cells with methylammonium lead halide semiconductors, a material that hardly anyone was studying just a few years ago. One hypothesis for this extraordinary performance has been suggested by Yanfa Yan and co-workers, who have performed density functional theory calculations and found that the defects likely to form in perovskites only create shallow traps that do not cause electrons and holes (positive charges) to rapidly recombine with each other¹⁰. Experiments still need to be performed to test their hypothesis, but the lessons learned through this study and others like it will help scientists continue to search for and design better materials for solar-energy conversion, whether they are for solar cells or solar water-splitting applications. With the theoretical and experimental tools available to us today, we need not be content with the photovoltaic technologies reported so far. Breakthroughs with perovskites over the past two years renew hope that better materials indeed are out there. □

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CHIRAL PLASMONIC NANOSTRUCTURES

Twisted by DNA

The optical properties of self-assembled plasmonic nanoparticles can be reversibly tuned by using DNA strands.

Andrea Di Falco

Noble-metal nanoparticles occupy the top drawer in the toolbox of scientists working with optical materials. These nanoparticles confine light in extremely small volumes, and their individual scattering properties can

be designed virtually at will by shaping and arranging them carefully. When their behaviour is considered collectively, the nanoparticles can also be used as building blocks (meta-atoms or metamolecules) for metamaterials — engineered materials

with effective homogeneous properties^{1,2}. In fact, the versatility of metamaterials is to a large degree determined by the ability to shape the units they are made from, and thus their collective properties. In particular, bottom-up fabrication methods based on