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本期梳理 2022 年 11 月 Nature、Science 期刊上材料科学领域的最新论文。





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材料科学

11月 Science 论文

[1]Initializing film homogeneity to retard phase segregation for stable perovskite solar cells 初始化薄膜均匀性以延缓稳定钙钛矿太阳能电池的相分离

出版信息: Science, 18 NOVEMBER 2022, VOL 378, ISSUE 6621

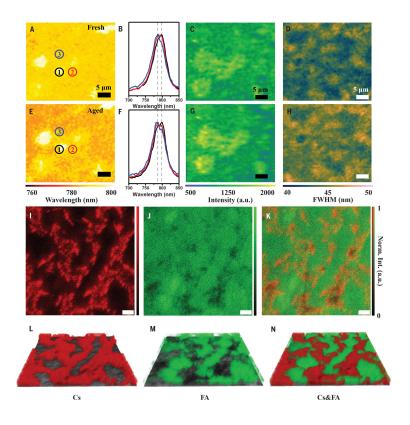
作者: YANG BAI, ZIJIAN HUANG, XIAO ZHANG, JIUZHOU LU, XIUXIU NIU, ZIWEN HE, ET AL.

第一作者单位: Beijing Key Laboratory of Construction Tailorable Advanced Functional Materials and Green Applications, MIIT Key Laboratory for Low-dimensional Quantum Structure and Devices, Experimental Center of Advanced Materials, School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, P. R. China.

国内相关报道: https://news.science.org/doi/10.1126/science.abn3148

Abstract: The mixtures of cations and anions used in hybrid halide perovskites for high-performance solar cells often undergo element and phase segregation, which limits device lifetime. We adapted Schelling's model of segregation to study individual cation migration and found that the initial film inhomogeneity accelerates materials degradation. We fabricated perovskite films (FA1 - xCsxPbI3; where FA is formamidinium) through the addition of selenophene, which led to homogeneous cation distribution that retarded cation aggregation during materials processing and device operation. The resultant devices achieved enhanced efficiency and retained >91% of their initial efficiency after 3190 hours at the maximum power point under 1 sun illumination. We also observe prolonged operational lifetime in devices with initially homogeneous FACsPb(Br0.13I0.87)3 absorbers.

摘要翻译: 用于高性能太阳能电池的杂化卤化物钙钛矿中使用的阴阳离子混合物经常发生元素和相分离,这限制了器件的使用寿命。研究组采用谢林偏析模型来研究单个阳离子的迁移,发现初始薄膜的不均匀性加速了材料的降解。通过添加硒酚制备钙钛矿薄膜(FA1-xCsxPbI3;其中 FA 为甲脒)可导致均匀的阳离子分布,延缓了材料加工和设备运行中的阳离子聚集。制备的器件效率大幅提升,在 1 倍太阳光照的最大功率点下运行3190 小时后,仍保持其初始效率的91%。研究组还观察到初始均质 FACsPb(Br0.13I0.87)3 吸收体延长了器件的使用寿命。



Mechanical nanolattices printed using nanocluster-based photoresists

使用基于纳米簇光刻胶印刷的机械纳米晶格

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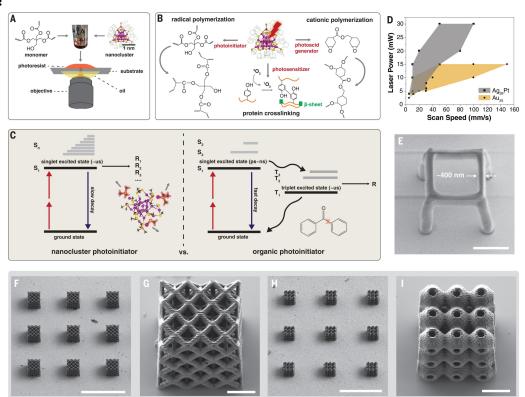
作者: QI LI, JOHN KULIKOWSKI, DAVID DOAN, OTTMAN A. TERTULIANO, CHARLES J. ZEMAN IV, MELODY M. WANG, ET AL.

第一作者单位: Department of Mechanical Engineering, Stanford University, Stanford, CA 94305, USA.

全文链接: https://www.science.org/doi/10.1126/science.abo6997

Abstract: Natural materials exhibit emergent mechanical properties as a result of their nanoarchitected, nanocomposite structures with optimized hierarchy, anisotropy, and nanoporosity. Fabrication of such complex systems is currently challenging because high-quality three-dimensional (3D) nanoprinting is mostly limited to simple, homogeneous materials. We report a strategy for the rapid nanoprinting of complex structural nanocomposites using metal nanoclusters. These ultrasmall, quantum-confined nanoclusters function as highly sensitive two-photon activators and simultaneously serve as precursors for mechanical reinforcements and nanoscale porogens. Nanocomposites with complex 3D architectures are printed, as well as structures with tunable, hierarchical, and anisotropic nanoporosity. Nanocluster-polymer nanolattices exhibit high specific strength, energy absorption, deformability, and recoverability. This framework provides a generalizable, versatile approach for the use of photoactive nanomaterials in additive manufacturing of complex systems with emergent mechanical properties.

摘要翻译: 天然材料因其具有优化的层次结构、各向异性和纳米孔隙率的纳米结构、纳米复合结构而呈现出新兴机械性能。目前制造这样复杂的系统颇具挑战性,因为高质量的三维(3D)纳米打印大多限于简单、均匀的材料。研究组报道了一种利用金属纳米簇快速纳米打印复杂结构纳米复合材料的策略。这些超小、量子受限的纳米簇可作为高度敏感的双光子激活剂,同时作为机械增强剂和纳米级致孔剂的前驱体。研究组已打印出具有复杂 3D 结构以及具有可调、分层和各向异性纳米孔隙率结构的纳米复合材料。纳米簇-聚合物纳米晶格具有高比强度、能量吸收、可变形性和可恢复性。该框架提供了一种可推广、通用的方法,有望实现光活性纳米材料应用于具有新兴机械性能的复杂系统增材制造。



Inhibiting creep in nanograined alloys with stable grain boundary networks 驰豫晶界网络显著抵抗金属高温蠕变

出版信息: Science, 11 November 2022, Volume 387 Issue 6620

作者: B. B. ZHANG, Y. G. TANG, Q. S. MEI, X. Y. LI AND K. LU

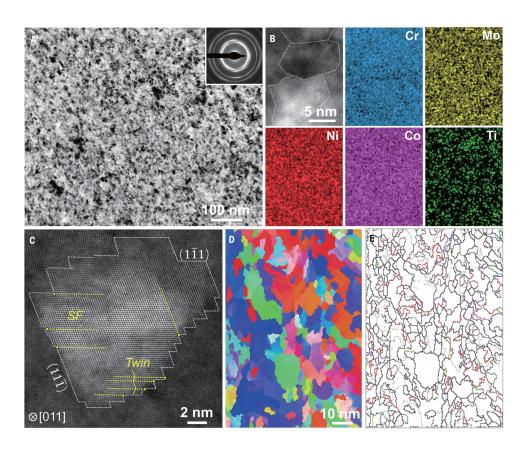
第一作者单位: Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China.

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全文链接: https://www.science.org/doi/10.1126/science.abq7739

Abstract: We report a different strategy to inhibiting creep by use of stable GB networks. Plastic deformation triggered structural relaxation of high-density GBs in nanograined single-phased nickel-cobalt-chromium alloys, forming networks of stable GBs interlocked with abundant twin boundaries. The stable GB networks effectively inhibit diffusional creep processes at high temperatures. We obtained an unprecedented creep resistance, with creep rates of ~10 - 7 per second under gigapascal stress at 700° C (~61% melting point), outperforming that of conventional superalloys.

摘要翻译:在此报告了一种通过使用驰豫的晶界网络来抑制蠕动的不同策略。塑性变形触发了纳米颗粒单相镍钴铬合金中高密度晶界的结构弛豫,形成了与丰富孪晶边界互锁的驰豫晶界网络。驰豫的晶界网络有效抑制了高温下的扩散蠕变过程。我们获得了前所未有的抗蠕变能力,在 700°C(约 61%熔点)、1GPa下,蠕变速率约为 10-7/s,优于传统高温合金。



材料科学

11月 Nature 论文

[1]Separating water isotopologues using diffusion-regulatory porous materials 利用扩散调节多孔材料分离水同位素

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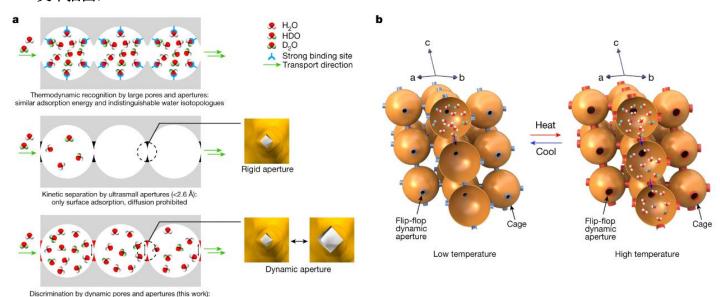
作者: Yan Su, Ken-ichi Otake, Jia-Jia Zheng, Satoshi Horike, Susumu Kitagawa & Cheng Gu

第一作者单位: State Key Laboratory of Luminescent Materials and Devices, Institute of Polymer Optoelectronic Materials and Devices, South China University of Technology, Guangzhou, P. R. China

全文链接: https://www.nature.com/articles/s41586-022-05310-y

Abstract: The discovery of a method to separate isotopologues, molecular entities that differ in only isotopic composition, is fundamentally and technologically essential but remains challenging. Water isotopologues, which are very important in biological processes, industry, medical care, etc. are among the most difficult isotopologue pairs to separate because of their very similar physicochemical properties and chemical exchange equilibrium. Herein, we report efficient separation of water isotopologues at room temperature by constructing two porous coordination polymers (PCPs, or metal – organic frameworks) in which flip-flop molecular motions within the frameworks provide diffusion-regulatory functionality. Guest traffic is regulated by the local motions of dynamic gates on contracted pore apertures, thereby amplifying the slight differences in the diffusion rates of water isotopologues. Significant temperature-responsive adsorption occurs on both PCPs: H2O vapour is preferentially adsorbed into the PCPs, with substantially increased uptake compared to that of D2O vapour, facilitating kinetics-based vapour separation of H2O/HDO/D2O ternary mixtures with high H2O separation factors of around 210 at room temperature.

摘要翻译: 开发一种分离同位素(仅同位素组成不同的分子实体)的方法从根本上和技术上都至关重要,但仍颇具挑战性。水同位素在生物过程、工业、医疗保健等方面非常重要,因其物理化学性质和化学交换平衡非常相似,水同位素是最难分离的同位素对之一。研究组报道了通过构建两种多孔配位聚合物(PCP,或金属-有机框架)在室温下高效分离水同位素的方法,其中框架内的翻折分子运动提供了扩散调节功能。流量受收缩孔隙上动态闸门的局部运动调节,从而放大了水同位素扩散速率的微小差异。两种 PCP 上都发生了显著的温度响应吸附: H2O 蒸汽优先被吸附到 PCP 中,与 D2O 蒸汽相比吸收量大幅增加,促进了 H2O/HDO/D2O 三元混合物基于动力学的蒸汽分离,在室温下 H2O 分离系数高达 210 左右。



A solution-processed n-type conducting polymer with ultrahigh conductivity

一种溶液处理的 n 型超高导电聚合物

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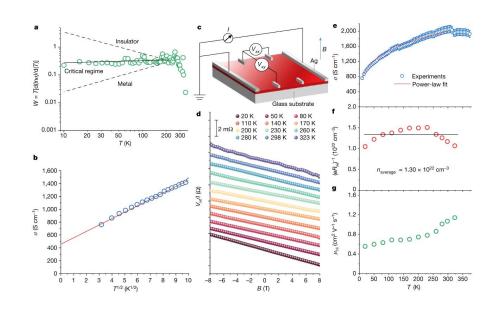
作者: Haoran Tang, Yuanying Liang, Chunchen Liu, Zhicheng Hu, Yifei Deng, Han Guo, et al.

第一作者单位: Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology (SCUT), Guangzhou, China

全文链接: https://www.nature.com/articles/s41586-022-05295-8

Abstract: Conducting polymers (CPs) with high conductivity and solution processability have made great advances since the pioneering work on doped polyacetylene, thus creating the new field of 'organic synthetic metals'. Various high-performance CPs have been realized, which enable the applications of several organic electronic devices. Nevertheless, most CPs exhibit hole-dominant (p-type) transport behaviour, whereas the development of n-type analogues lags far behind and only a few exhibit metallic state, typically limited by low doping efficiency and ambient instability. Here we present a facilely synthesized highly conductive n-type polymer poly(benzodifurandione) (PBFDO). The reaction combines oxidative polymerization and in situ reductive n-doping, greatly increasing the doping efficiency, and a doping level of almost 0.9 charges per repeating unit can be achieved. The resultant polymer exhibits a breakthrough conductivity of more than 2,000 S cm-1 with excellent stability and an unexpected solution processability without extra side chains or surfactants. Furthermore, detailed investigations on PBFDO show coherent charge-transport properties and existence of metallic state. The benchmark performances in electrochemical transistors and thermoelectric generators are further demonstrated, thus paving the way for application of the n-type CPs in organic electronics.

摘要翻译: 自掺杂聚乙炔的开创性工作以来,具有高导电性和溶液可加工性的导电聚合物(CPs)取得了巨大进展,从而开创了"有机合成金属"的新领域。各种高性能 CP 的实现使多种有机电子器件的应用成为可能。尽管如此,大多数 CP 表现为空穴主导(p型)输运行为,而 n 型类似物的发展远远滞后,很少表现出金属态,通常受到低掺杂效率和环境不稳定性的限制。研究组介绍了一种易于合成的高导电性 n 型聚合物聚(苯并二呋喃二酮)(PBFDO)。该反应结合了氧化聚合和原位还原 n 掺杂,大大提高了掺杂效率,每重复单元可达到几乎 0.9 电荷的掺杂水平。所得聚合物具有超过 2000 S cm-1 的突破性电导率,兼具优异的稳定性和出乎意料的溶液可加工性,且无需额外的侧链或表面活性剂。此外,对 PBFDO 的详细研究表明了相干电荷输运特性和金属态的存在。研究组进一步证明了电化学晶体管和热电发电机的基准性能,为 n 型 CP 在有机电子领域的应用铺平了道路。



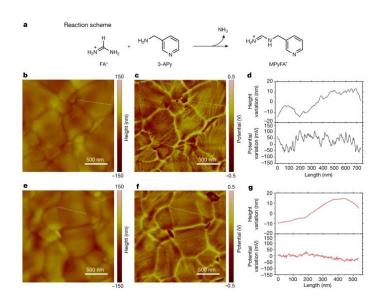
Surface reaction for efficient and stable inverted perovskite solar cells 高效稳定倒置钙钛矿太阳能电池的表面反应

出版信息: Nature, 10 November 2022, VOL 611, ISSUE 7935

作者: Qi Jiang, Jinhui Tong, Yeming Xian, Ross A. Kerner, Sean P. Dunfield, Chuanxiao Xiao, et al. 第一作者单位: Chemistry and Nanoscience Center, National Renewable Energy Laboratory, Golden, CO, USA 全文链接: https://www.nature.com/articles/s41586-022-05268-x

Abstract: Perovskite solar cells (PSCs) with an inverted structure (often referred to as the p-i-n architecture) are attractive for future commercialization owing to their easily scalable fabrication, reliable operation and compatibility with a wide range of perovskite-based tandem device architectures. However, the power conversion efficiency (PCE) of p-i-n PSCs falls behind that of n-i-p (or normal) structure counterparts. This large performance gap could undermine efforts to adopt p-i-n architectures, despite their other advantages. Given the remarkable advances in perovskite bulk materials optimization over the past decade, interface engineering has become the most important strategy to push PSC performance to its limit. Here we report a reactive surface engineering approach based on a simple post-growth treatment of 3-(aminomethyl)pyridine (3-APy) on top of a perovskite thin film. First, the 3-APy molecule selectively reacts with surface formamidinium ions, reducing perovskite surface roughness and surface potential fluctuations associated with surface steps and terraces. Second, the reaction product on the perovskite surface decreases the formation energy of charged iodine vacancies, leading to effective n-type doping with a reduced work function in the surface region. With this reactive surface engineering, the resulting p-i-n PSCs obtained a PCE of over 25 per cent, along with retaining 87 per cent of the initial PCE after over 2,400 hours of 1-sun operation at about 55 degrees Celsius in air.

摘要翻译:具有倒置结构(通常称为 p-i-n 结构)的钙钛矿太阳能电池(PSCs)由于其易于扩展的制造、可靠的操作以及与广泛钙钛矿串联器件结构的兼容性,对未来的商业化颇具吸引力。然而,p-i-n PSCs 的功率转换效率(PCE)低于 n-i-p(或正常)结构的同类产品。尽管 p-i-n 体系结构有其他优势,但这种巨大的性能差距可能会削弱采用该体系结构的努力。鉴于过去十年中钙钛矿块体材料优化取得的显著进展,界面工程已成为将 PSC 性能推向极限的最重要策略。研究组报道了一种基于钙钛矿薄膜顶部 3-(氨甲基)吡啶(3-apy)的简单生长后处理的反应性表面工程方法。首先,3-APy 分子选择性地与表面甲酰胺离子反应,降低钙钛矿表面粗糙度和与表面步/阶地相关的表面电位波动。第二,钙钛矿表面的反应产物降低了带电碘空位的形成能,导致表面区域功函数降低的有效 n 型掺杂。通过这种反应性表面工程,产生的 p-i-n PSCs 获得了超过 25%的 PCE,并在大约 55℃的空气中 1-太阳光照下运行超过 2400 小时后,仍保留了 87%的初始 PCE。



Ultra-bright, efficient and stable perovskite light-emitting diodes

超亮、高效、稳定的钙钛矿发光二极管

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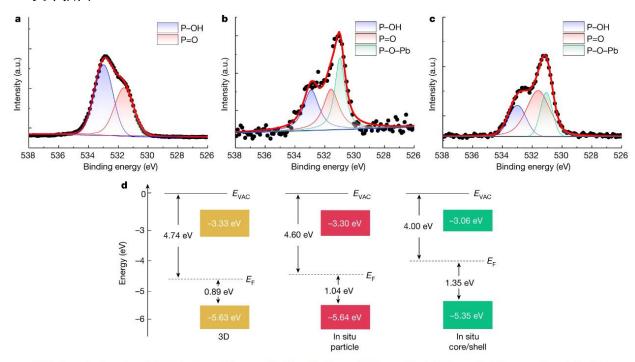
作者: Joo Sung Kim, Jung-Min Heo, Gyeong-Su Park et al.

第一作者单位: Department of Materials Science and Engineering, Seoul National University, Seoul, Republic of Korea

全文链接: https://www.nature.com/articles/s41586-022-05304-w

Abstract: Here, we report an ultra-bright, efficient and stable PeLED made of core/shell perovskite nanocrystals with a size of approximately 10 nm, obtained using a simple in situ reaction of benzylphosphonic acid (BPA) additive with three-dimensional (3D) polycrystalline perovskite films, without separate synthesis processes. During the reaction, large 3D crystals are split into nanocrystals and the BPA surrounds the nanocrystals, achieving strong carrier confinement. The BPA shell passivates the undercoordinated lead atoms by forming covalent bonds, and thereby greatly reduces the trap density while maintaining good charge-transport properties for the 3D perovskites. We demonstrate simultaneously efficient, bright and stable PeLEDs that have a maximum brightness of approximately 470,000 cd m-2, maximum external quantum efficiency of 28.9% (average = 25.2 ± 1.6% over 40 devices), maximum current efficiency of 151 cd A-1 and half-lifetime of 520 h at 1,000 cd m-2 (estimated half-lifetime >30,000 h at 100 cd m-2).

摘要翻译:报告一种由核/壳型钙钛矿纳米晶体制成的超亮、高效且稳定 PeLED,尺寸约为 10 nm,使用苄膦酸 (BPA)添加剂与三维多晶钙钛矿薄膜的简单原位反应,而不需要单独的合成过程。在反应过程中,大型三维晶体分裂为纳米晶体,BPA 包围纳米晶体,实现了强载流子限制。BPA 壳通过形成共价键钝化欠配位的铅原子,从而大大降低了陷阱密度,同时保持了三维钙钛矿良好的电荷输运特性。我们同时展示了高效、明亮和稳定的 PeLEDs,其最大亮度约为 47 万 cd m-2,最大外部量子效率为 28.9%(在 40 个器件中平均= 25.2± 1.6%),最大电流效率为 151 cd A-1,在 1000 cd m-2 下的半衰期为 520 h(估计在 100 cd m-2 下的半衰期为> 30000 h)。



 \mathbf{a} - \mathbf{c} , O 1s XPS core-level spectra of BPA (\mathbf{a}), in situ particle perovskite (\mathbf{b}) and in situ core/shell perovskite (\mathbf{c}). \mathbf{d} , Energy level diagram of the pristine 3D, in situ particle, in situ core/shell perovskites obtained from parameters derived from UPS spectra. (\mathcal{E}_{VAC} , vacuum level; \mathcal{E}_{F} , Fermi level).

Open-channel metal particle superlattices

开放通道金属粒子超晶格

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作者: Yuanwei Li, Wenjie Zhou, Ibrahim Tanriover et al.

第一作者单位:

Department of Chemical and Biological Engineering, Northwestern University, Evanston, IL, USA International Institute for Nanotechnology, Northwestern University, Evanston, IL, USA

全文链接: https://www.nature.com/articles/s41586-022-05291-y

Abstract: Here, a universal approach for synthesizing metallic open-channel superlattices with pores of 10 to 1,000 nm from DNA-modified hollow colloidal nanoparticles (NPs) is reported. By tuning hollow NP geometry and DNA design, one can adjust crystal pore geometry (pore size and shape) and channel topology (the way in which pores are interconnected). The assembly of hollow NPs is driven by edge-to-edge rather than face-to-face DNA – DNA interactions. Two new design rules describing this assembly regime emerge from these studies and are then used to synthesize 12 open-channel superlattices with control over crystal symmetry, channel geometry and topology.

摘要翻译: 本文报道一种用 DNA 修饰的空心胶体纳米粒子(NPs)、合成孔洞为 10~1000 nm 的金属开放管道式超晶格的通用方法。通过调整空心胶体纳米粒子几何结构和 DNA 设计,可以调整晶体孔隙几何结构(孔隙大小和形状)和通道拓扑结构(孔隙相互连接的方式)。空心胶体纳米粒子的构建是由边缘到边缘的 DNA-DNA 相互作用驱动的,而不是面对面的 DNA-DNA 相互作用。我们引入了两个新设计规则去描述这种构建过程,并合成了 12 个控制晶体对称性、通道几何和拓扑的开放管道式超晶格。

