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Bespoke crystalline hybrids towards the next generation of white LEDs

Authors: Jiawei Chen[1,†], Soumya Mukerjee[2,3,†], Weijin Li[1,2,*], Haibo Zeng[1,*], Roland A. Fischer[2,*]

[1] Institute of Optoelectronics & Nanomaterials, MIIT Key Laboratory of Advanced Display Materials and Devices & School of Materials Science and Engineering, Nanjing University of Science and Technology, Xiaolingwei Street 200, 210094, Nanjing, China. Email: zeng.haibo@njust.edu.cn; wjli@njust.edu.cn

[2] Chair of Inorganic and Metal-organic Chemistry, Catalysis Research Center, Ernst-Otto-Fischer Straße 1 and Department of Chemistry, Technical University of Munich, Lichtenbergstraße 4, 85748 Garching b. München, Germany. E-mail: roland.fischer@tum.de

[3] Bernal Institute, Department of Chemical Sciences, University of Limerick, Limerick, Ireland.

[†]J.C. and S.M. contributed equally.

Thanks to the lifespan and efficiency benchmarks set by the current generation of white light-emitting diodes (WLEDs), the lighting industry is quickly replacing traditional LEDs that use monochromatic light. Building upon recent research advances in framework solids for WLEDs, and capitalizing on their bottom-up design principles, modular crystalline hybrids are paving new paths to energy-efficient lighting alternatives.

As analysed in the World Energy Outlook 2021, light energy consumes approximately 20% of the world's electricity demand and accounts for 2.4% of its energy expenditure. It is thus responsible for about 12% of the global CO_2 emission footprint. Light-emitting diodes (LEDs) are electricity-driven light sources that substantially mitigate energy consumption. Their energy footprint is about 80% lower than that of traditional incandescent or fluorescent illuminators, a feature in direct alignment with the United Nations Sustainable Development Goal 7 of affordable and clean energy. As a testament to the success of white LEDs (WLEDs), half of the global lighting sector in 2019 was credited to WLEDs, and the 2030 market forecasts WLEDs to notch 90% of the full energy-pie.

In regard to luminous efficiency, WLEDs are handicapped by secondary energy loss. Besides, it offers luminous flux lower than incandescent and fluorescent illuminators. Moreover, bound by the mechanism of white light generation, colour rendering index (CRI) of WLEDs (~ 75%) falls short of incandescent illuminators (100%). Put simply, improving energy efficiency and lighting quality are the most crucial bottlenecks in advancing WLEDs. In pursuit of a select few top-performers likely to penetrate the market, of particular importance is the on-demand, bottom-up design of white light-emitting materials, or exploring novel mechanisms of white light generation.

White photoluminescence

Traditional LEDs generate white light using complementary lights. A blue or a UV LED chip is combined with a phosphor that partially absorbs blue or UV electroluminescence, converting the light to white photoluminescence (PL) (**Fig. 1a**).

Today's WLED technology relies on a blue LED component, gallium nitride (GaN). Although the early-1990s discovery of GaN won the Nobel Prize in Physics for revolutionizing the societal impact of WLEDs and revamping household appliances, GaN's brittleness and poor user-friendliness have made the material cost-inefficient[1]. The blue LED component also affords a high proportion of blue light in the resulting white light spectrum, dwindling the CRI and intensifying the blue light pollution that contributes to our sleep disorders and retinal damage. That an average person today spends more than 12 hours daily in front of illuminated screens makes the case for improving LEDs stronger than ever.

In 2014, halide perovskites were introduced as a new class of crystalline RGB (red, green and blue)-light-emitting materials[2]. Acting as the phosphor component, these perovskites filled in the missing colours in the RGB spectrum, improving the CRI over traditional phosphor-based WLEDs. Moreover, solution processability and structural flexibility render halide perovskites more versatile.

From WLEDs to white electroluminescence

White EL-based WLEDs outperform the traditional white PL-based, phosphor-converted WLEDs by overcoming secondary energy loss and simplifying device structures. Free from a blue light source, white EL is also primed to minimize blue light pollution.

White EL can be produced by combining multiple RGB LED chips, or by a single-source emitter (Fig. 1b). The RGB approach often suffers from self-absorption and colour separation and requires device engineering. These issues exacerbate benefit-to-cost ratios and largely restrict the use of RGB LEDs to micro- and mini-LEDs in indicators and displays.

Perovskite single-source WLED emitters, introduced in 2014, overcome these shortcomings in principle[3]. In single-source emitting perovskites, self-trapped excitons (STEs) cause lattice distortion, broadband emission and large Stokes shift. Acting as phosphor, STE-laced perovskites can be activated by UV chips to elicit white PL, with PL quantum yields up to 86%[4]. Nevertheless, poor charge transport properties prevented efficient white EL in STE perovskite materials, until a 2021 study introduced STEs in lead halide perovskites[5]. In that work, a cesium lead iodide (α/δ-$CsPbI_3$) perovskite monolayer-based WLED delivered a maximum luminance of 12,200cdm^{-2} and an external quantum efficiency of 6.5%, surpassing the threshold values for commercial lighting (luminance of ~10^3–10^4 cdm^{-2}, external quantum efficiency of ~6%). Not only do these $CsPbI_3$ newcomers have a wealth of merits – earth-abundant raw materials, low-cost fabrication, high energy conversion efficiency, excellent recyclability and the organic unity of excellent charge transport properties from α-$CsPbI_3$ and STE broadband emission from δ-$CsPbI_3$ – of particular relevance is their de novo design. For molecular WLEDs laced with STEs, the right choice of building blocks is key to boosting carrier mobility. We believe this success to be the first step towards adopting bespoke crystalline hybrids in WLED applications.

By-design crystalline hybrids

Crystallinity is all important in WLED design, as it influences carrier recombination, charge extraction, transport, diffusion length and electrical stability. Regardless, translating new crystals into electroluminescent devices will become moot if the main problem of weak charge transport (an effect of insulating nature) remains unsolved. Weak charge transport can be an outcome of redox-inactive ligands and/or frail electronic interactions between the metal centres and ligands. Modular framework solids, thanks to their amenability to crystal engineering, can overcome these[6].

Hybridizing single-source electroluminescent perovskite emitters with crystalline framework solids – such as the compositionally modular metal–organic frameworks (MOFs), covalent organic frameworks (COFs) and metal–organic cages — can intuitively leverage performance benchmarks and, ergo, fast-track the market penetration of electroluminescent emitters[7,8] (Fig. 1c). Across the several families of perovskites, growth regulation, controlled surface modification and passivating structural defects are likely to improve performance and device stability. Modular crystalline solids can regulate these processes. It is also unlikely that there will be a 'one size fits all' STE-laced crystalline hybrid, given the interdependence between structural forms, experimental conditions and optimized performance metrics. Among the advantages of modular crystalline solids is that their properties can be tuned by design, through structure and composition. For instance, by leveraging reticular chemistry principles, electrical conductivity of a layered nickel MOF delivered record-high performance[9,10]. We envision that amalgamating single-source white electroluminescent emitters (predicated upon STEs) with conductive crystalline hybrids will overcome the instability and poor conductivity of STE-based perovskites, and push the boundaries of lighting science and technology. Materials scientists must capitalize on the advantages of conductive MOF and COF hybrids to take these bespoke materials beyond lab-scale demonstration, inching closer to commercial adoption.

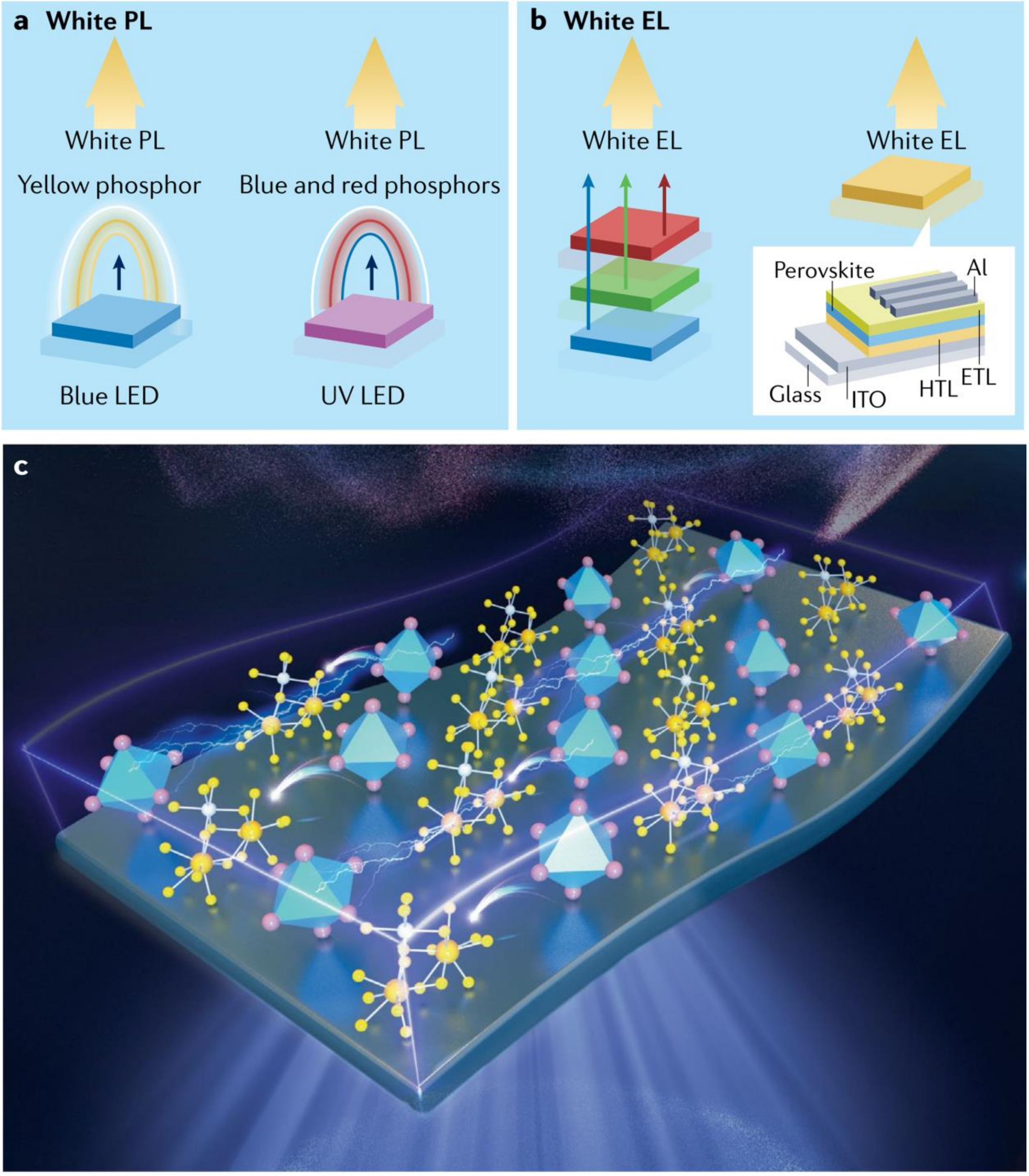

Fig. 1 | Models of white light emission. a | White photoluminescence (PL) is generated using a blue or UV LED chip with phosphors that emit yellow or blue and red light, respectively. **b** | White electroluminescence (EL) is generated by combining red, green and blue LED chips, or by single-source electroluminescent white light emitters based on self-trapped excitons. **c** | A bespoke white-light-emitting crystalline hybrid, combining the concepts of modular framework solids and self-trapped exciton-based perovskites. ETL, electron transport layer; HTL, hole transport layer; ITO, indium-doped tin oxide.

Competing interests

The authors declare no competing interests.

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