Efficient light-emitting diodes based on oriented perovskite nanoparticles

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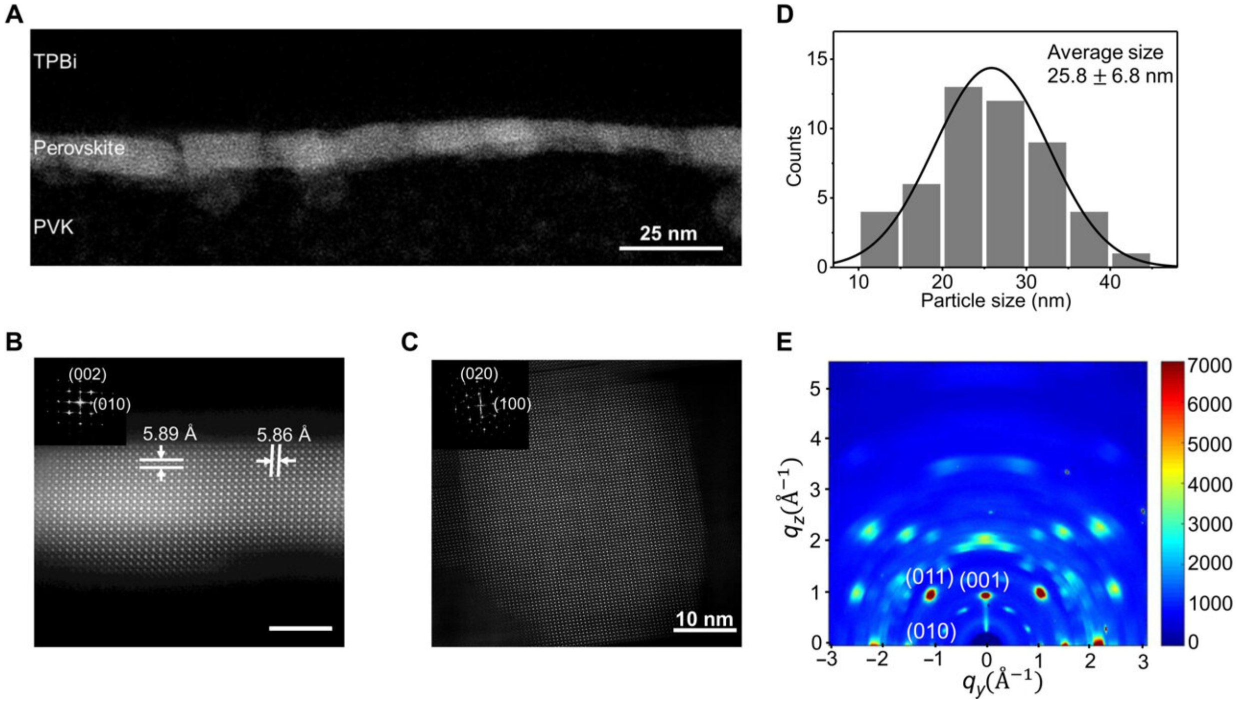


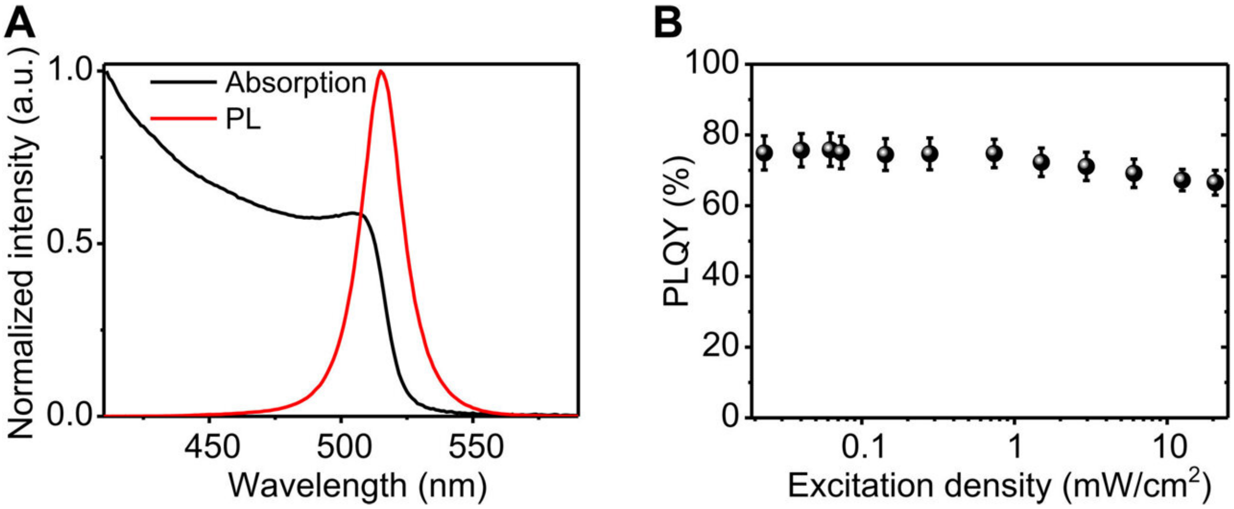
Structural characterizations of the perovskite nanoplatelet films. (A) A cross-sectional scanning transmission electron microscopy–high-angle annular dark-field (STEM-HAADF) image showing the continuous and pinhole-free perovskite layer. TPBi, 2,2′,2″-(1,3,5-benzinetriyl)tris(1-phenyl-1H-benzimidazole); PVK, poly(9-vinylcarbazole). (B) A zoomed-in STEM-HAADF image showing the fine structure of a perovskite nanoplatelet. Inset: The corresponding fast Fourier transform (FFT) pattern. (C) A typical high-resolution transmission electron microscopy (HRTEM) image of the perovskite nanoplatelets dispersed on a copper grid. Inset: The corresponding FFT pattern. (D) Statistical diagram of the size distribution of the nanoplatelets measured by HRTEM. The average size is 25.8 nm and the corresponding SD is 6.8 nm. The

Gaussian fitting is provided as a guide to the eye. (E) Grazing-incidence wide-angle x-ray scattering pattern. The diffraction spots originate from the crystal faces of nanoplatelets. The two diffraction spots at qz = 1.065 and qy = 1.070 Å−1 correspond to {001} and {010} of β-$CsPbBr_3$, respectively. Credit: *Science Advances*, 10.1126/sciadv.abg8458

Planar perovskite light-emitting diodes (LEDs) are high-performance and cost-effective electroluminescence devices that are ideal for large-area display and lighting applications. By exploring the emission layers with high ratios of horizontal transition dipole moments (TDMs), researchers can boost the photon outcoupling of planar LEDs. The LEDs that are based on anisotropic perovskite are inefficient due to the challenges of regulating the orientations of TDMs as well as the difficulties of achieving high photoluminescence quantum yields, including challenges of realizing charge balance in the films of assembled nanostructures. In this work, Jieyuan Cui and a research team in chemistry, materials science and optics in China, showed efficient electroluminescence emanating from an in-situ perovskite film made of a monolayer of nanoplatelets. The team achieved LEDs with a peak external quantum efficiency (EQE) of 23.6 percent to represent highly efficient planar perovskite LEDs.

Transition dipole moments and metal halide perovskites

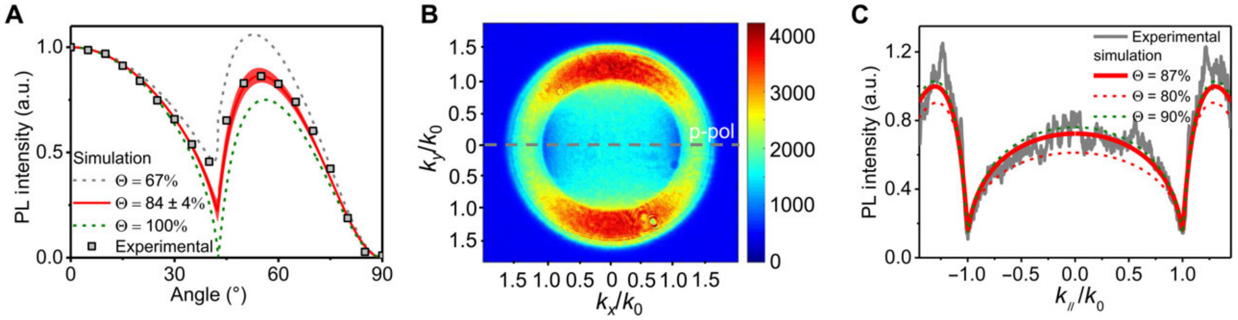
The photon emission characteristics in semiconductors are based on transition dipole moments. Molecules in a material can attain an excited or non-excited state through the absorption and emission of light, where the rules of transition dipole moment and quantum mechanics can help predict if the transition to an excited state is likely. Nanoplatelets and nanorods that comprise optical transition dipole moments within materials are highly anisotropic and their structure-property relationship is of interest for planar light-emitting diodes (LEDs). Generally, transition dipole moments are horizontally oriented for light coupling and those that are vertically oriented contribute to energy loss. Metal halide perovskites are another emerging class of solution-processed-semiconductors with interesting properties including high photoluminescence quantum yields and tunable emission wavelengths. In this report, Cui et al. described efficient LEDs based on in situ grown perovskite films to show high ratios of horizontal transition dipole moments and high photoluminescence quantum yields.



Optical properties of the perovskite nanoplatelet films. (A) Absorption and PL (excited by a 405-nm laser) spectra. a.u., arbitrary units. (B) Excitation intensity–dependent PLQY. The error bars represent the experimental uncertainties in the PLQY measurements at 0.4 mW/cm2 and the errors in the determination of relative PL intensities and excitation power. Credit: *Science Advances*, 10.1126/sciadv.abg8458

Structural characterization of nanoplatelets

The device contained a perovskite layer analyzed by aberration-corrected scanning transmission electron microscopy (STEM). The team deposited the perovskite film from a precursor solution containing several compounds including lithium bromide, cesium bromide and lead bromide dissolved in dimethyl sulfoxide (DMSO). Thereafter, using high-angle annular dark-field (HAADF) images, Cui et al observed a smooth perovskite film. Using zoom-in studies they noted well-resolved atom columns with highly crystalline perovskite nanoplatelets. Thereafter, using atomic force microscopy, they determined the roughness of the material and understood the size of the perovskite crystals or nanoplatelets using high-resolution transmission electron microscopy.



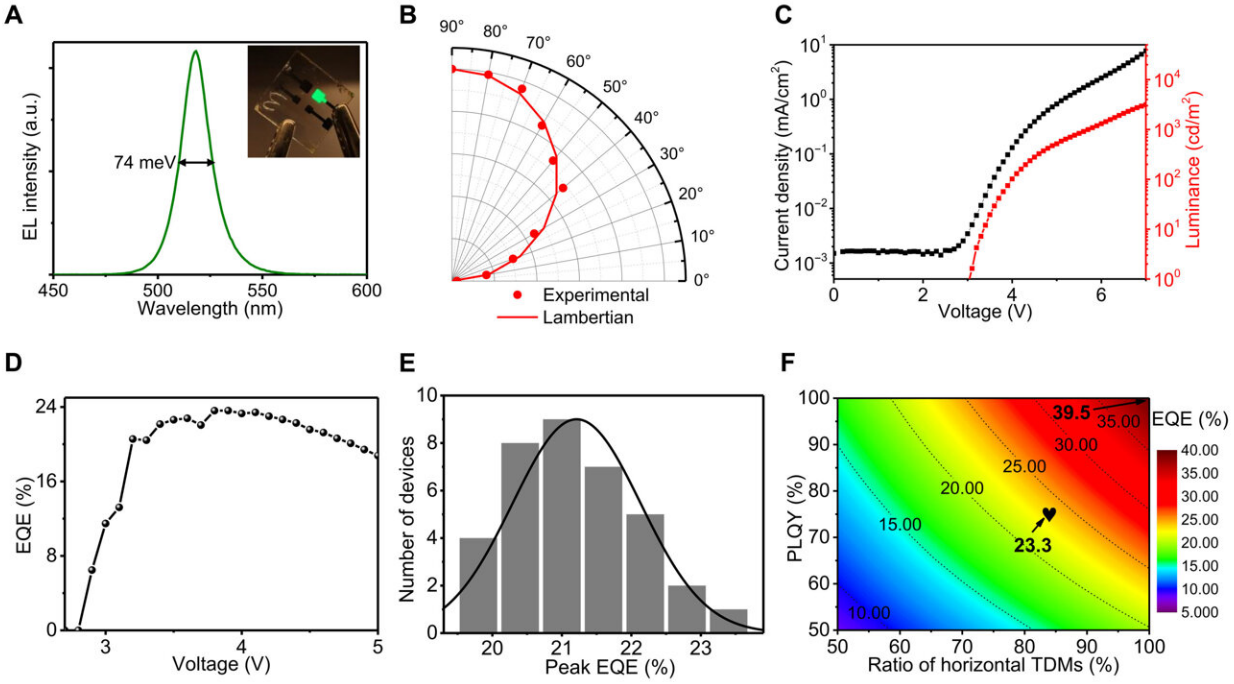
Orientations of the TDMs of the perovskite nanoplatelet films. (A) Angle-dependent PL measurements of the perovskite film on a quartz/TFB/PVK substrate. The experimental data (gray squares) are fitted by the classical electromagnetic dipole model (red line), giving a horizontal TDM ratio of 84 ± 4%. (B) Back focal plane (BFP) image of a perovskite film. (C) p-polarized line cut (gray line) along the dashed line in of the BFP image (B). This line cut is fitted with a horizontal TDM ratio of 87% (red solid line). Credit: *Science Advances*, 10.1126/sciadv.abg8458

Optical analyses of the nanoplatelet film

The team influenced the electronic and optical properties of the perovskite film using the quantum confinement effect and then quantified the orientation of transition dipole moments of the perovskite film. Thereafter, Cui et al. analyzed the light emission of the perovskite film using black focal plane (BFP) spectroscopy. To accomplish this, they probed a small region of the perovskite nanoplatelet film with a laser for photoexcitation. The data indicated excellent spatial uniformity of the horizontal orientation of transition dipole moments in the film. The team next used the BFP data of four spots from different regions to show excellent spatial uniformity of the orientations of horizontal transition dipole moments in the films. Due to the concentration of the bulky organic ammonium cations and the presence of lithium bromide in the precursor solution, the perovskite nanoplatelet film oriented with high photoluminescence quantum yields. By doubling the concentration of the bulky organic ammonium cations, Cui et al. formed perovskite films with strong excitonic absorption peaks and credited the horizontal orientation of the nanoplatelets on the flat substrates to Van der Waals interactions.

Characterizing the room temperature–operating perovskite LEDs

Based on further experiments, the team showed how the introduction of lithium bromide (LiBr) in the precursor solution improved the photoluminescence quantum properties of the film. Additionally, the electroluminescence spectrum of the perovskite nanoplatelet film indicated ultrapure green emissions and the pin-hole free morphology of the nanoplatelet film allowed negligible current leakage. When they performed optical simulations on the materials by using the classical dipole model developed for planar microcavities, the results indicated high-outcoupling efficiency of 31.1 percent for the perovskite devices based on the orientation of the nanoplatelet film. While previous work aimed to control the orientations of transition dipole moments by focusing on the assembly of anisotropic colloidal nanostructures, high-efficiency electroluminescence required the syntheses of anisotropic colloidal nanostructures with high quantum yield. The potential to fulfill the device requirements were challenging due to material design and assembly requirements.



Device characterizations of the green LEDs based on the perovskite nanoplatelet films. (A) EL spectrum. Inset: Photograph of an operating green LED (effective area: 3.24 mm2). (B) Angular distribution of the EL intensity follows the Lambertian profile. (C) Current density–luminance–voltage characteristics of a typical device. (D) EQE-voltage relationship of the device with a champion EQE of 23.6%. (E) Histogram of peak EQEs from 36 devices. The Gaussian fits are provided as a guide to the eye. (F) Contour plot of the simulation results of device EQE as a function of PLQY and Θ of the perovskite emissive layer. The device structure shown in (A) is used for the simulation. The refractive indexes of the multilayers are obtained by ellipsometer. For our perovskite nanoplatelet film with a PLQY of ~75% and a Θ of 84%, the optical simulation predicts a maximum EQE of ~23.3%. Credit: *Science Advances*, 10.1126/sciadv.abg8458

Outlook

In this way, Jieyuan Cui and colleagues showed how the orientation of transition dipole moments of perovskite films could be regulated to overcome the limits of light-outcoupling of planar LEDs to form green LEDs with exceptionally high external quantum efficiency of up to 23.6 percent. The chemical versatility of the perovskite materials allowed Cui et al. to extend the facile approach to in situ grown nanoplatelet films to develop differently colored LEDs with high external quantum efficiency . The work describes a simple and effective method to understand the role of the anisotropic optical properties of nanostructures in the formation of optoelectronic devices.

More information: Jieyuan Cui et al, Efficient light-emitting diodes based on oriented perovskite nanoplatelets, *Science Advances* (2021). DOI: 10.1126/sciadv.abg8458

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Brandon R. Sutherland et al, Perovskite photonic sources, *Nature Photonics* (2016). DOI: 10.1038/nphoton.2016.62

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