Imaging bandgaps of perovskite solar cells with luminescence

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We develop a non-destructive, camera-based luminescence method to capture bandgap images of perovskite solar cells (PSCs) with high spatial resolution. This new technique utilizes the well-defined and symmetric band-to-band luminescence spectrum emitted from perovskite materials, whose spectral peak locations present their bandgaps. We apply the technique to image the bandgap variation across various high-efficiency perovskite solar cells, and the bandgap non-uniformity within the same device due to material degradation. We also verify our results with micro-photoluminescence (µ-PL) spectroscopy scans. The agreement between the two techniques opens exciting opportunities for this imaging concept to be a quantified, high spatially-resolved, large-area characterization tool of PSCs. This invention continues to fortify the high value of luminescence imaging for the research and development of this novel photovoltaic technology.

Method descriptions

In general, a band-to-band luminescence spectrum emitted from a semiconductor can be described in the form of 

$$I(\lambda) = A \times f(u_1, u_2, ..., u_N)$$

where $A$ is an intensity scaling factor, $\{u_1, u_2, ..., u_N\}$ are $N$ unknowns, and $f$ is an arbitrary function operator describing spectral shapes. In luminescence images, emitted photons pass through various optical components before reaching a detector. The detected intensity in each pixel is a wavelength-dependent integration of the product of the emitted spectrum and optical responses of system components located along the pathway between the sample and the detector. Generally, we have the following relationship between the detected intensity $I$ and the emitted spectrum $I(\lambda)$ from the sample:

$$I = \int I(\lambda) \times R_{det}(\lambda) \times T_{objective}(\lambda) \times T_{filters}(\lambda) d\lambda$$

(Eq.1)

$R_{det}$ is the spectral response of the detector, $T_{objective}$ is the transmission response of the objective lens used to image the sample, and $T_{filters}$ is the combined transmission response of all optical filters. These parameters are all wavelength dependent and their values are known for a certain imaging system. Therefore, it is possible to extract all unknowns $\{u_1, u_2, ..., u_N\}$ from the intensity $I$ with enough equations. In this way, it is analogous to solving $N$ unknowns using $N$ equations. A simple way to achieve this is to capture luminescence intensity images with various optical filters having different transmissivities.

Results

Highly-efficient organic-inorganic hybrid PSCs have been reported to yield symmetrical band-to-band PL spectral shapes at room temperature without any sub-bandgap signal. To verify it, we capture PL spectra from various perovskite films with different compositions (Fig. 1). We observe that these spectra can be reasonably well fitted with Gaussian distributions in which the peak location ($\lambda_0$) and full-width at half-maximum (FWHM) are two fitting parameters. This peak location ($\lambda_0$) is considered as the bandgap of perovskite materials. These observations allow us to simplify Eq.1 into:

$$I = A \times \int Gaussian(\lambda_0, FWHM) \times R_{det}(\lambda) \times T_{objective}(\lambda) \times T_{filters}(\lambda) d\lambda$$

(Eq.2)
Thus, if we capture 3 luminescence intensity images using 3 different optical filters, two intensity ratios among the 3 images will cancel out the pre-factor $A$, and leave us with $\{\lambda_0, \text{FWHM}\}$. We can simulate these two ratios for a range of $\{\lambda_0, \text{FWHM}\}$ and pre-store the data. They will serve as a look-up table to extract the peak wavelength (i.e. the bandgap) of perovskite materials.

Fig. 1: PL spectra of various perovskite films and corresponding Gaussian fits. (a) MAPbI$_3$. (b) FA$_{0.85}$MA$_{0.15}$PbI$_{2.55}$Br$_{0.45}$. (c) Cs$_{0.05}$Rb$_{0.05}$FA$_{0.765}$MA$_{0.135}$PbI$_{2.55}$Br$_{0.45}$. MA = methylammonium, FA = formamidinium.

Fig. 2a shows an electroluminescence (EL) image of a degraded PSC. Applying our method presented above, we can convert this image into a bandgap image (Fig. 2b). The results are confirmed with a μ-PL spectroscopy map (Fig. 2c). This allows correlating the degraded regions with their bandgap shifting (Fig. 2d). This finding cannot be revealed with a standard EL/PL image.

Fig. 2: (a) EL intensity image of a degraded perovskite solar cell. (b) Extracted bandgap image from the EL intensity image. (c) Bandgap image extracted from the micro-PL scan, confirming the results from the EL imaging method. (d) Peak location (i.e. bandgap) versus EL intensity.

The final presentation will provide an in-depth underlying physics of this novel technique and demonstrate it on various high-efficiency PSCs. This fast, non-destructive luminescence-based technique could be a powerful tool to characterize large-area PSCs, especially with the rapid upscale of this promising solar cell technology.