Highly Luminescent and Stable Quasi-2D Perovskites based on Multi-functional Asymmetric Spacer

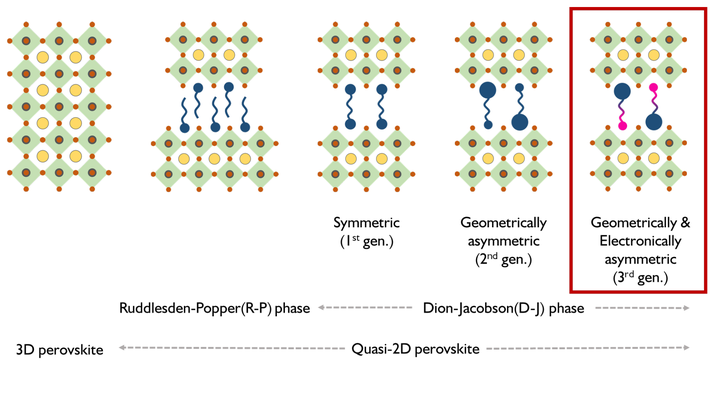
MinJae Kim[1], Seungmin Shin[1], Himchan Cho[1*]

[1] Department of Materials Science and Engineering, KAIST, Daejeon, KAIST
* E-mail : himchan@kaist.ac.kr

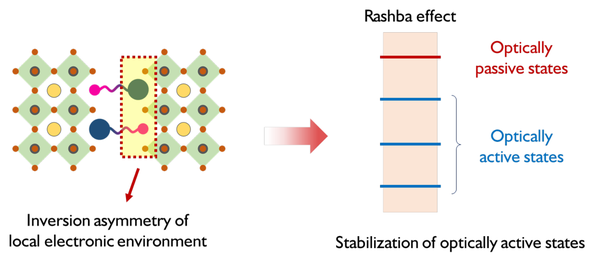
KAIST NGON Next-Generation Optoelectronic Nanomaterials Lab.

Introduction

Dion-Jacobson phase perovskites

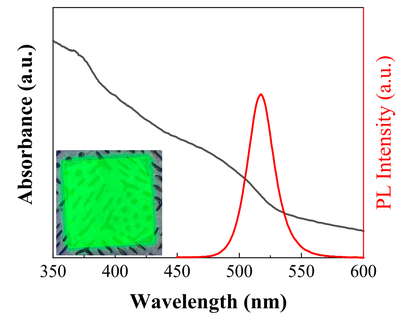


Collapse of inversion symmetry



- **Dion-Jacobson perovskites hold good stability and carrier conductivity, whilst photoluminescence quantum yield(PLQY) remains inferior**
- **Such foible can be surmounted with the electronically asymmetric spacer**

Highly Emissive & Stable Quasi-2D perovskite



- ✓ PLQY > 50%(on PEDOT:PSS:PFI)
- ✓ FWHM < 25 nm

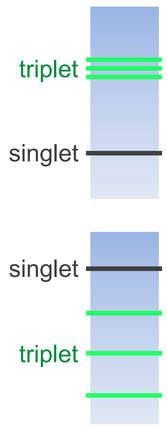
Theoretical

Inversion symmetry held

$$E_{0,0} = \Lambda + K$$

$$E_{0,(0,\pm1)} = \Lambda - K$$

$$K = \langle \psi_e(1)\psi_h(2) | \widehat{H}_{Coulomb} | \psi_e(2)\psi_h(1) \rangle < 0$$



Inversion symmetry collapsed

$$\widehat{H_{R,z}} = \sum_{c=e,h} \left(\alpha_{xy}^{z,c} \sigma_x^c \sigma_y^c - \alpha_{yx}^{z,c} \sigma_y^c \sigma_x^c \right), \quad et\ cycl.$$

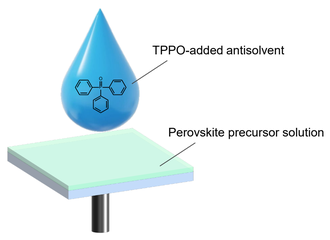
$$E = \Gamma \left(\pm \alpha_{xy}^{z,e} \alpha_{xy}^{z,h} \pm \alpha_{yx}^{z,e} \alpha_{yx}^{z,h} \right)$$

Experimental

Quasi-2D perovskite film fabrication

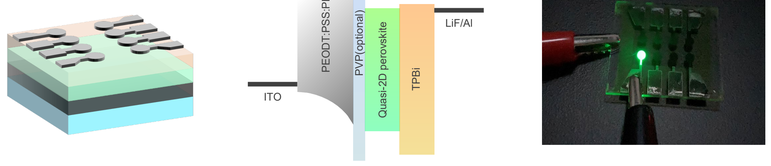
Fabrication condition

- Spin-coating of perovskite precursor solution onto the substrate
- Antisolvent dripping as designated time
- Passivation agent: TPPO



- **Quasi-2D perovskite film was successfully fabricated on the substrate**
- **The film was annealed at 70℃ for 10 min to remove any residual solvents**

PeLED Fabrication



- **Hole transport layer and emission layer were solution-processed**
- **TPBi(50nm), LiF(1nm), and Al(100nm) was deposited via thermal evaporation**

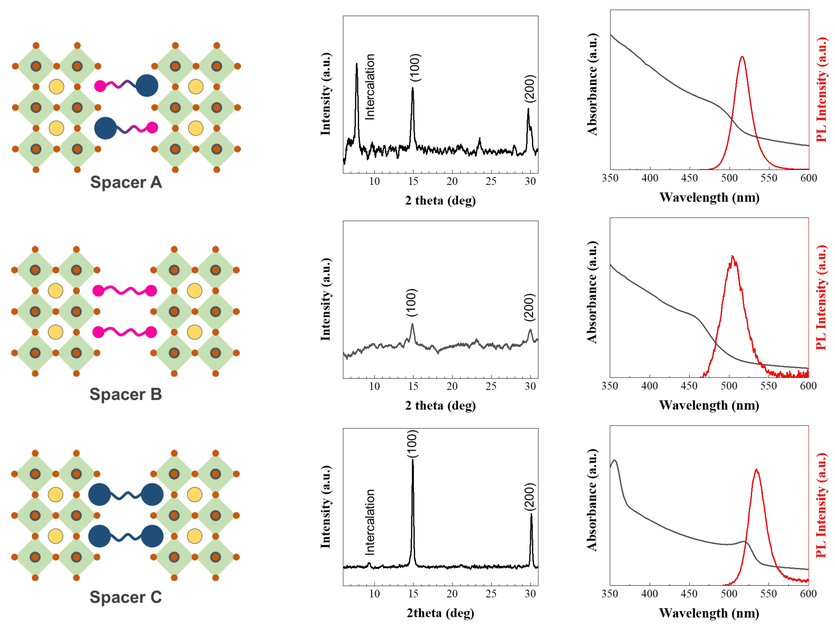
Conclusion

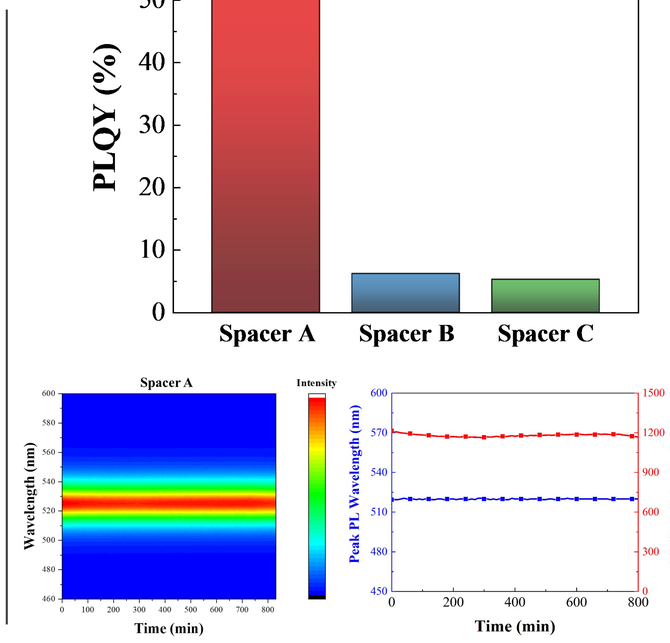
- **Here, the heretofore underrated aspect of Dion-Jacobson phase perovskite, the electrical asymmetry of the spacer was demonstrated.**
- **Emission enhancement cannot be attributed to the passivation effect and perovskite slab alignment.**
- **Therefore, the improvements are attributed to the electronic inversion asymmetry of the spacer molecule, which stabilizes optically active states relative to the passive state, thereby enabling bright emission.**
- **Light-emitting diodes based on the quasi-2D perovskite emission layer were fabricated and recorded greatly enhanced EQE, luminance, and color purity.**
- **Additional optimization in both electroluminescence and photoluminescence will be further pursued.**

This research was supported by the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (NRF-2022M3H4A1A03085346).

Results

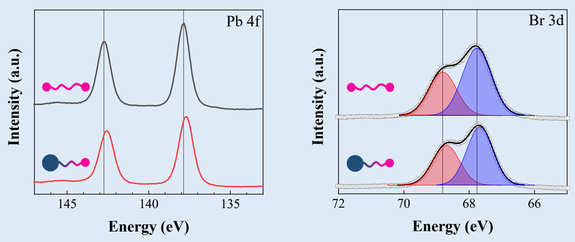
Optical properties upon spacer inversion symmetry modulation





Hypothesis 1

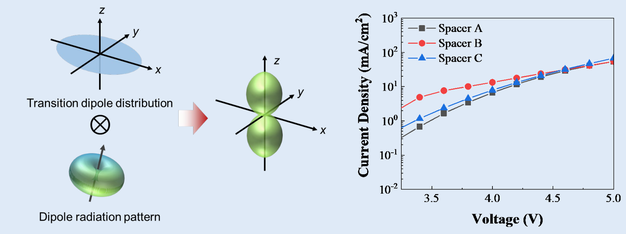
- **Enhancement of PLQY is attributable to the passivation capability of the bulky head of the spacer molecule**



- **XPS survey revealed that the bulky head of the spacer passivates Pb^{2+} to some extent**
- **But such a passivation effect seems not salient, because if so, spacer C should have the highest PLQY → Reject Hypothesis 1**

Hypothesis 2

- **Enhancement of PLQY is attributable to the horizontal alignment of the quasi-2D perovskite slabs**

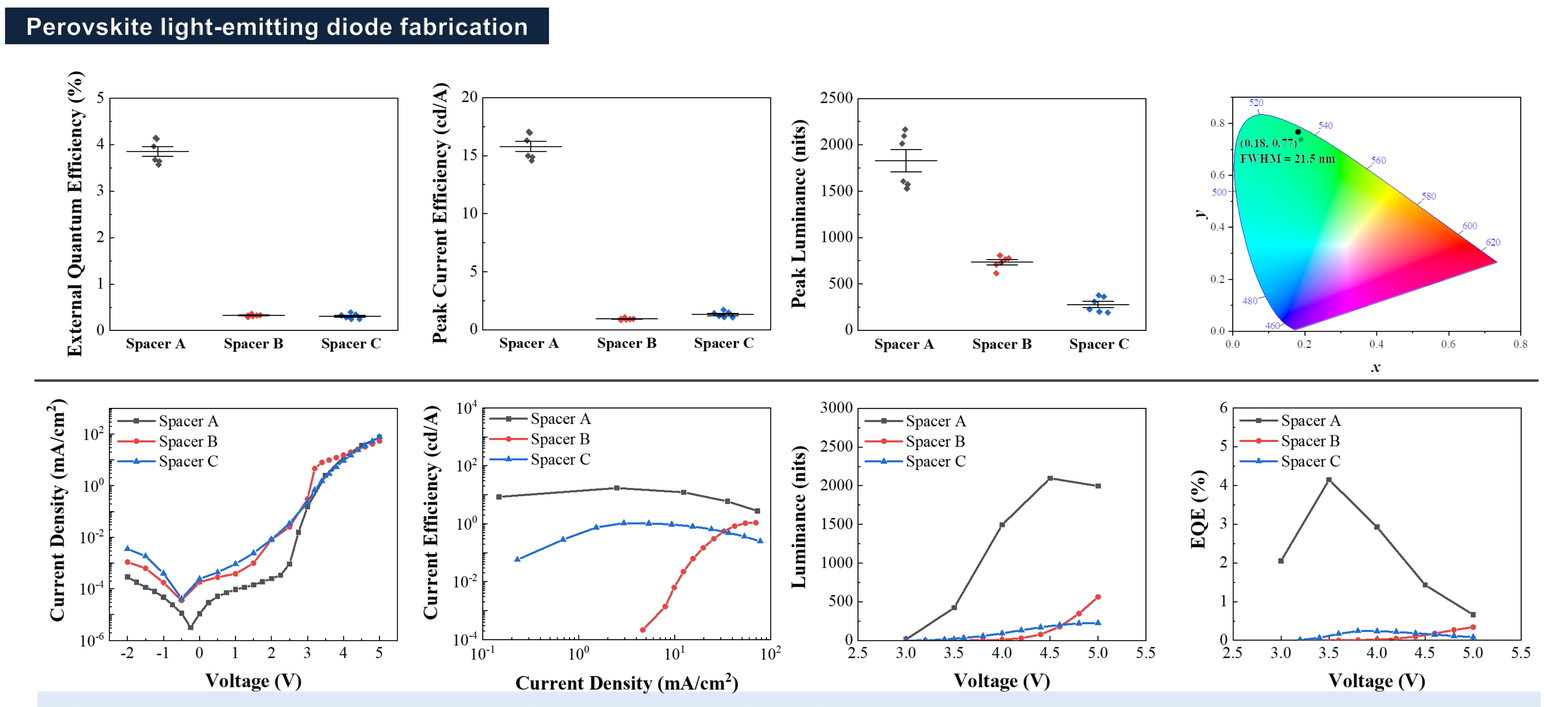


- **Consistent with *J-V* characteristics in which spacer A case shows the lower current density**
- **But such behavior cannot explain PL characteristics that spacer A shows higher PL intensity→ Reject Hypothesis 2**

→ Therefore, optical enhancement is attributed to the electrical asymmetry of the spacer molecule

→ The collapse of inversion symmetry of local electric potential stabilizes optically active states

Perovskite light-emitting diode fabrication



- **Quasi-2D perovskite light-emitting diodes were fabricated, recording EQE > 4%, luminance > 2000 nits with narrow FWHM ~ 21.5 nm**
- **Electroluminescence characteristics were superior in the case of the asymmetric spacer, in consensus with photoluminescence characteristics**
- **Ideality factor η over 2 implies layered structure was successfully formed**