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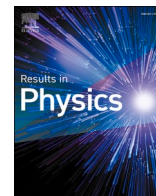
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Microcavities integrated in metal halide perovskite light-emitting field-effect transistors

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ABSTRACT

Metal halide perovskites are materials that show unique characteristics for photovoltaics and light emission. Amplified spontaneous emission and stimulated emission has been shown with these materials, together with electroluminescence in light-emitting diodes and light-emitting transistors. An important achievement that combine stimulated emission and electroluminescence could be the fabrication of electrically driven metal halide perovskite lasers.

In this work, the integration of metal halide perovskite light-emitting field-effect transistors with photonic microcavities is proposed. This can lead to the engineering of electrically driven lasers. The microcavities have been designed in order to have the cavity mode at 750 nm, which is the peak wavelength of the electroluminescent spectrum of recently reported MaPbI_3 -based electroluminescent devices. The optical properties of the photonic microcavities have been simulated by means of the transfer matrix method, considering the wavelength dependent refractive indexes of all the materials involved. The material for the gate is indium tin oxide, while different materials, either inorganic or organic, have been considered for the microcavity architectures.

Introduction

Metal halide perovskites show ground-breaking properties in terms of photovoltaic efficiency and light emission [1]. In April 2021, a power conversion efficiency of 25.6 % has been reached with a perovskite solar cell [2]. In July 2022, 31.25 % efficiency for tandem perovskite-silicon solar cell has been achieved [3]. Concerning light emission, metal halide perovskite based light emitting diodes are very established [4,5]. Furthermore, amplified spontaneous emission and laser emission has been observed in many reports with metal halide perovskites [6–8]. The very recent review by Liu et al. [9] reports different types of optically pumped metal halide perovskite laser cavities, such as whispering gallery mode (WGM), Fabry-Pérot, vertical cavity surface emitting laser (VCSEL). Outstanding laser performances have been achieved, with very low lasing threshold. It is noteworthy that a CsPbBr_3 quantum dot based VCSEL has shown laser emission at 522 nm with a linewidth of 0.9 nm and an ultralow lasing threshold of $0.39 \mu\text{J}/\text{cm}^2$ [10]. With MAPbI_3 thin films, laser emission at longer wavelengths has been achieved. For example, with a MAPbI_3 thin film based VCSEL, laser emission has been shown at 778 nm, with a linewidth of 0.2 nm and a laser threshold of $7.6 \mu\text{J}/\text{cm}^2$ [11]. The combination of laser emission and electroluminescence is very challenging, but it can lead to the fabrication of electrically

driven perovskite lasers, a breakthrough for many applications from photonics to communications. Interesting structures are proposed to achieve such device [12].

The use of light-emitting field-effect transistors could be strategic for electrically driven lasing, since they allow a high carrier density together with the control of current flow, charge injection and emission patterns [13,14]. The ambipolar characteristics of metal halide perovskites, such as methylammonium lead iodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$ or MAPbI_3), allow the fabrication of MAPbI_3 -based light-emitting transistors [15–17]. The integration of a laser cavity in light-emitting diodes could be pursued via the inclusion of photonic crystals in the transistors. Organic light-emitting transistors with photonic crystal gate dielectrics have been proposed in previous reports [18,19], demonstrating electroluminescence spectral modulation and enhancement.

In this work, we have designed different microcavities integrated with light-emitting field effect transistors based on metal halide perovskites. These architectures could allow the fabrication of electrically driven metal halide perovskite lasers in which the microcavities provide the feedback mechanism. Different materials are employed to widen the fabrication possibilities: inorganic materials such as silicon dioxide, titanium dioxide, fluorine indium co-doped cadmium oxide (FICO), silicon; organic polymers as polyvinyl carbazole (PVK), cellulose acetate

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(CA). The transfer matrix method has been employed considering all the wavelength dependent refractive indexes of the materials.

Methods

In this work the transmission spectra of the microcavities have been simulated with the transfer matrix method, a well established tool to study one dimensional photonic structures [20,21]. Within this method the matrix used for the k th layer is given by

$$M_k = \begin{bmatrix} \cos\left(\frac{2\pi}{\lambda}n_k(\lambda)d_k\right) & -\frac{i}{n_k(\lambda)}\sin\left(\frac{2\pi}{\lambda}n_k(\lambda)d_k\right) \\ -in_k(\lambda)\sin\left(\frac{2\pi}{\lambda}n_k(\lambda)d_k\right) & \cos\left(\frac{2\pi}{\lambda}n_k(\lambda)d_k\right) \end{bmatrix} \quad (1)$$

with $n_k(\lambda)$ the wavelength dependent refractive index of the layer and d_k the thickness of the layer (in nm). The matrix describing the whole multilayer system is given by the product of the M_k matrices

$$M = \prod_{i=1}^N M_k = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \quad (2)$$

The integer number N represents the number of layers. From the matrix elements of the matrix M it is possible to compute the transmission coefficient

$$t = \frac{2n_s}{(M_{11} + M_{12}n_0)n_s + (M_{21} + M_{22}n_0)} \quad (3)$$

and, consequently, the transmission

$$T = \frac{n_0}{n_s}|t|^2 \quad (4)$$

In Eqs. (4) and (5), n_0 and n_s are the refractive indexes of air and glass, respectively.

For silicon dioxide, the following Sellmeier equation has been employed [22,23]

$$n_{SiO_2}^2(\lambda) - 1 = \frac{0.6961663\lambda^2}{\lambda^2 - 0.0684043^2} + \frac{0.4079426\lambda^2}{\lambda^2 - 0.1162414^2} + \frac{0.8974794\lambda^2}{\lambda^2 - 9.896161^2} \quad (5)$$

For titanium dioxide, the wavelength-dependent refractive index is given by [24]

$$n_{TiO_2}(\lambda) = \left(4.99 + \frac{1}{96.6\lambda^{1.1}} + \frac{1}{4.60\lambda^{1.95}}\right)^{1/2} \quad (6)$$

For PVK, the Sellmeier equation for the refractive index of is given by [25,26]:

$$n_{PVK}^2(\lambda) - 1 = \frac{0.09788\lambda^2}{\lambda^2 - 0.3257^2} + \frac{0.6901\lambda^2}{\lambda^2 - 0.1419^2} + \frac{0.8513\lambda^2}{\lambda^2 - 1.1417^2} \quad (7)$$

For CA, the Sellmeier equation for the refractive index of CA is [25,26]:

$$n_{CA}^2(\lambda) - 1 = \frac{0.6481\lambda^2}{\lambda^2 - 0.0365^2} + \frac{0.5224\lambda^2}{\lambda^2 - 0.1367^2} + \frac{2.483\lambda^2}{\lambda^2 - 13.54^2} \quad (8)$$

In the Eqs. (5) to (8) λ is in micrometers.

To predict the behaviour of the plasmonic response in our photonic structures the Drude model can be employed [27,28], where the frequency dependent complex dielectric function of ITO and FICO can be written as:

$$\epsilon_{FICO}(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega) \quad (9)$$

where

$$\epsilon_1 = \epsilon_\infty - \frac{\omega_p^2}{(\omega^2 - \Gamma^2)} \quad (10)$$

and

$$\epsilon_2 = \frac{\omega_p^2\Gamma}{\omega(\omega^2 - \Gamma^2)} \quad (11)$$

with

$$\omega_p = \sqrt{\frac{Ne^2}{\epsilon_0 m^*}} \quad (12)$$

For ITO $N = 2.49 \times 10^{26} \text{ cm}^{-3}$, $\epsilon_\infty = 4$, $m^* = 0.4m_e$ and $\Gamma = 0.1132 \text{ eV}$ [29]. For FICO $N = 1.68 \times 10^{27} \text{ cm}^{-3}$, $\epsilon_\infty = 5.6$, $m^* = 0.43m_e$ and $\Gamma = 0.07 \text{ eV}$ [29,30].

The wavelength dependent refractive index of silicon is taken from Ref. [31]. Considering the studied wavelength range for the microcavity that includes silicon layers (500 – 1500 nm), the imaginary part of the refractive index is neglected. Finally, the wavelength dependent refractive of the active material MAPbI₃ is taken from Refs. [32,33]. Such refractive index dispersion includes the real part and the imaginary part.

Results and discussion

The typical structure of a light-emitting transistor integrated with a microcavity is depicted in Fig. 1, in agreement with the structures fabricated in Refs. [18,19]. As gate contact, indium tin oxide has been selected following the transistor design reported in Ref. [19]. Such material is interesting for light-emitting devices because of its transparency in the visible range. The source and drain contacts are placed at the two sides of the MaPbI₃ layer. Several materials can be used as source and drain, such as gold/nickel [15] or gold/titanium [34].

For all the microcavities studied in this work, the selected thickness for the MaPbI₃ layer is 40 nm, as reported in the fabrication of a MAPbI₃-based light-emitting diode [5]. To tune the thickness of the defect, the MaPbI₃ layer is embedded between two layers of another material. In the example in Fig. 1, the two one dimensional photonic crystals are made of titanium dioxide and FICO, while the MaPbI₃ layer is embedded between two layers of silicon dioxide. The employment of FICO is interesting since it could allow also the possibility of UV light-induced dielectric function change via photodoping [30].

In Fig. 2 the transmission spectrum of the structure depicted in Fig. 1 is shown. The one-dimensional photonic crystals have six bilayers of TiO₂ and FICO. The cavity mode with the highest transmission is centred at 750 nm, where previously reported MAPbI₃-based light-emitting diodes show the peak wavelength of the electroluminescent spectrum

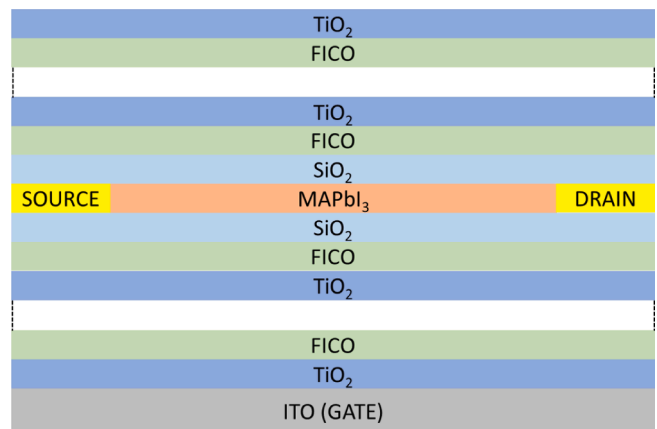


Fig. 1. Structure of the MaPbI₃ light-emitting field-effect transistor integrated with a microcavity composed by two FICO/TiO₂ one-dimensional photonic crystals with the MaPbI₃ layer as defect.

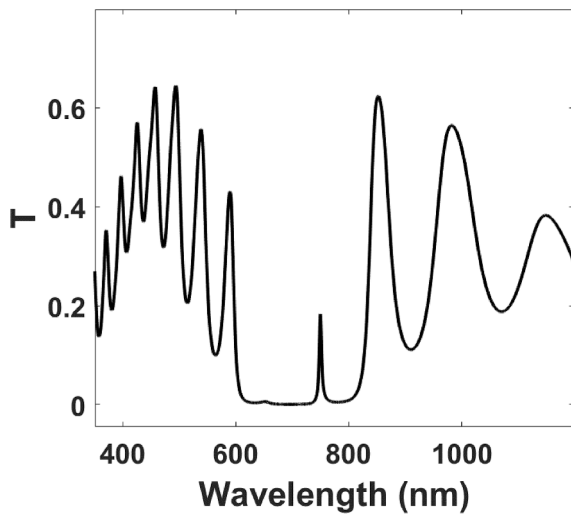


Fig. 2. Transmission spectrum of ITO/(TiO₂/FICO)₆/SiO₂/MaPbI₃/SiO₂/(FICO/TiO₂)₆.

[5,12]. The microcavity shows an additional cavity mode at 652 nm, very weak in terms of transmission.

The thickness of the ITO layer is 100 nm. The thickness of the titanium dioxide layers is 73.5 nm, while the thickness of the FICO layers is 112.7 nm. With 6 bilayers of TiO₂ and FICO, the total thickness of each photonic crystal is 1117 nm. In order to have the cavity mode at 750 nm, the MaPbI₃ layer is embedded between two layers of silicon dioxide with a thickness of 144.2 nm. In this structure, the weaker defect mode at 652 nm is more evident.

Since the photonic crystal between the ITO-based gate and the active material, i.e. MaPbI₃, has to act as a gate dielectric, its thickness is crucial for the operation of the light-emitting transistor. Moreover, also the thickness silicon dioxide buffer layer within the cavity should be considered.

The thickness can be controlled with the number of bilayers in the photonic crystals. With 4 bilayers of TiO₂ and FICO, the total thickness of each photonic crystal is 745 nm (Fig. 3), in line with the thickness employed in a previously reported light-emitting transistor [19].

A microcavity in which the one-dimensional photonic crystal is made of SiO₂ and FICO and the defect is made of TiO₂/MaPbI₃/TiO₂ shows a transmission spectrum as in Fig. 4. The thickness of the silicon dioxide

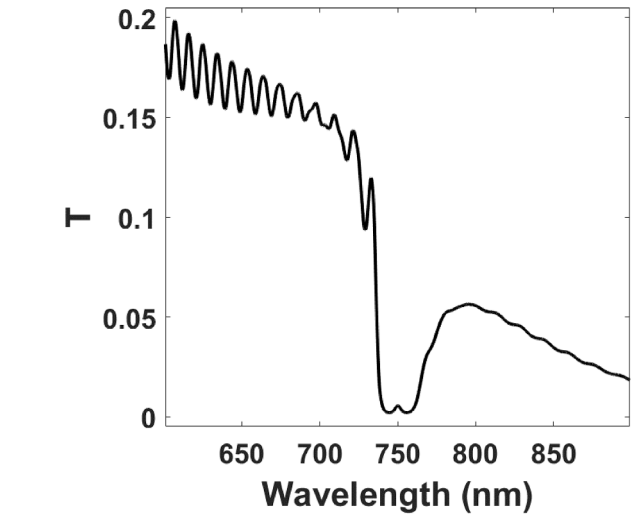


Fig. 4. Transmission spectrum of ITO/(SiO₂/FICO)₅₀/TiO₂/MaPbI₃/TiO₂/(FICO/SiO₂)₅₀.

layers is 128.4 nm, while the thickness of FICO is 124.1 nm. The thickness of the titanium dioxide layers is 139.5 nm. Because of the similar real part of the refractive index of silicon dioxide and FICO, the photonic band gap is relatively narrow and a high number of bilayers is needed to achieve a sufficiently efficient photonic band gap. Because of the high number of layers of FICO the transmission strongly decreases in the near infrared.

To achieve very thin photonic crystals, and thus a very thin gate dielectric ITO and MaPbI₃, a structure alternating silicon dioxide and silicon is designed (Fig. 5). The thickness of the silicon layers is 53.1 nm, while the thickness of the silicon dioxide layers is 123.9 nm. The thickness of the titanium dioxide layers in the microcavity defect is 139.5 nm. Thus, the total thickness of the dielectric between ITO and MaPbI₃ is 493.5 nm. This thickness of 493.5 nm is very promising for the realization of high performing metal halide light-emitting transistors, since such value is in line with the dielectric thicknesses reported in Refs. [15–17].

Finally, a microcavity with fully organic photonic crystals has been designed, allowing completely different fabrication techniques such as spin coating (Fig. 6) [35]. The two one-dimensional photonic crystals are made of PVK and CA. With these two materials, the refractive index

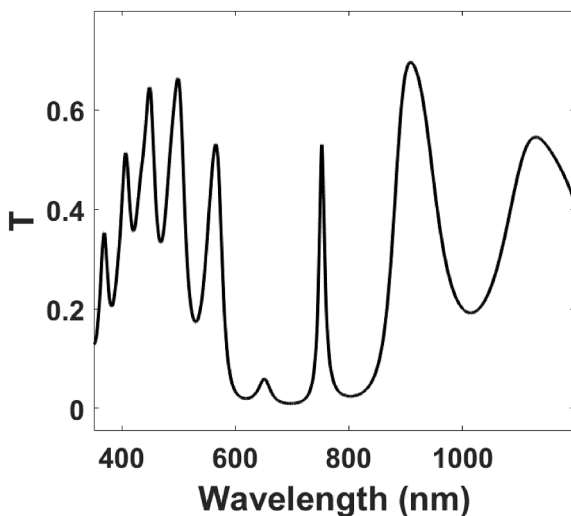


Fig. 3. Transmission spectrum of ITO/(TiO₂/FICO)₄/SiO₂/MaPbI₃/SiO₂/(FICO/TiO₂)₄.

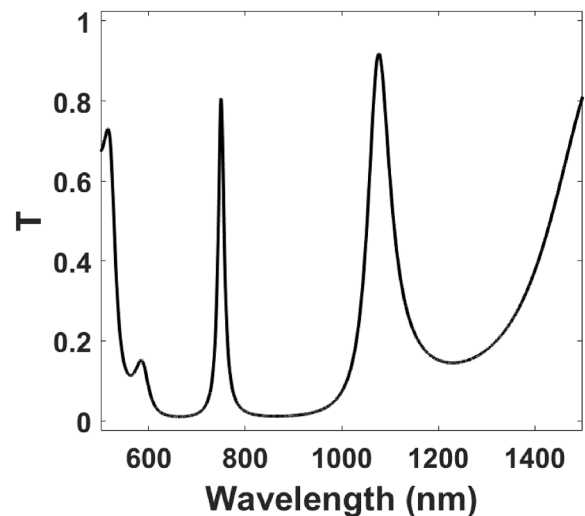


Fig. 5. Transmission spectrum of ITO/(SiO₂/Si)₂/TiO₂/MaPbI₃/TiO₂/(Si/SiO₂)₂.

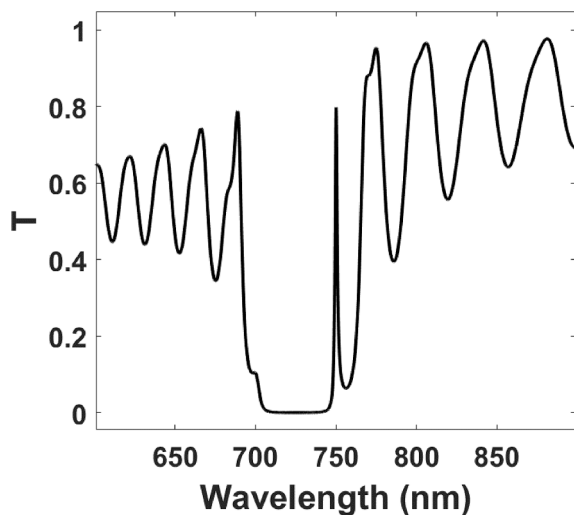


Fig. 6. Transmission spectrum of ITO/(CA/PVK)₂₅/CA/MaPbI₃/CA/(PVK/CA)₂₅.

is quite high for polymers. However, to achieve an efficient photonic band gap, a high number of bilayers is needed.

Conclusion

Different microcavities integrated with metal halide based light-emitting field-effect transistors have been designed by means of transfer matrix method. Different materials have been employed in order to provide a library of diverse architectures. The architectures include ITO as gate dielectric and the active material MaPbI₃ is embedded between two one-dimensional photonic crystals. The photonic crystals have been made with silicon, silicon dioxide, titanium dioxide, FICO, PVK and CA. With the couple Si/SiO₂, very thin photonic crystals have been engineered. These architectures, that are compatible with MAPbI₃-based light-emitting transistors [15–17] and MAPbI₃-based VCSELs [11] reported in literature, could allow the fabrication of electrically driven metal halide perovskite lasers.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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