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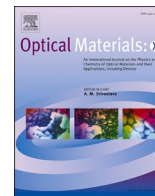
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MaPbI₃ and 2D hybrid organic-inorganic perovskite based microcavities employing periodic, aperiodic, and disordered photonic structures with light-induced tuning possibility

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ABSTRACT

Inorganic-organic perovskites semiconductors are becoming increasingly interesting due to their remarkable optical properties, such as a high photoluminescence quantum yield and the possibility to show optical gain in a broad range of wavelengths. We have here simulated microcavities that embed MaPbI₃ and 2D hybrid organic-inorganic perovskite semiconductors by sandwiching such active layers between periodic, aperiodic and disordered photonic structures. The complex refractive index dispersions of MaPbI₃ and 2D hybrid organic-inorganic perovskite have been recently reported in literature. Thus, we have carefully considered the refractive index dispersion of all the materials employed, such as silicon dioxide, titanium dioxide, and the aforementioned perovskite layers. Moreover, by employing a photochromatic polymer, namely the diarylethene-based polyester pDTE, we have designed a microcavity with light-induced tuning of the cavity modes is possible.

1. Introduction

Organic-inorganic halide perovskite semiconductors have become in few years highly important in photonics due to their impressive nonlinear optical properties [1–4]. These materials are interesting because of their unique performances as photovoltaic materials, but also because of their high photoluminescence quantum yield and optical gain, making them promising for light-emitting diodes and lasers [5]. Just to cite a few examples, perovskite-based lasers have been monolithically integrated into a silicon nitride photonic circuit [6] and perovskite-based random lasing with cavity exciton-polaritons has been reported [7].

In photonics, the simplest building block is the photonic crystal [8–10], characterized by the alternation of different refractive indexes in one, two, or three directions. The one-dimensional photonic crystals are particularly interesting because they can be fabricated with a large variety of techniques [11], the optical properties can be studied with high accuracy [12], both for enhancement of spontaneous emission [13, 14] and lasers [15–19]. One-dimensional photonic structures can be designed by following aperiodic and disordered (random) patterns.

These structures can be exploited to tune and enhance the optical characteristics of emitting materials embedded in such structures [20–23].

One-dimensional photonic crystals that include perovskites have been used to fabricate efficient solar cells with tunable structural colour [24]. Moreover, microcavities based on polymeric materials that embed perovskites have been designed and fabricated [25].

For a proper design of the photonic structures including perovskite semiconductors as a constituent material, it is necessary to know the real part and the imaginary part of the refractive index as a function of the wavelength, i.e. the complex refractive index dispersion. The refractive index of MAPbI₃ has been reported by several groups in Refs. [26–29]. Recently, also the refractive indexes of 2D hybrid organic-inorganic perovskite (HOIP) semiconductors in Ruddlesden–Popper (RP) and Dion–Jacobson (DJ) phases have been reported [30].

In this work, we have exploited the determination of the complex refractive index dispersion of these perovskite materials. With such dispersions we have simulated photonic microcavities, that embed MaPbI₃ and 2D hybrid organic-inorganic perovskite semiconductors, by sandwiching such active layers between periodic, aperiodic, and

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disordered photonic structures. We demonstrate the possibility to observe cavity modes with the different structures. Moreover, we demonstrate the possibility to obtain a tunable microcavity by adding layers of diarylethene-based polyester (pDTE) in the microcavity.

2. Methods

We have used the transfer matrix method [31–33] to simulate the light transmission spectra of the differently designed microcavities.

In this study, we consider a system glass/multilayer/air that is impinged by the light with normal incidence.

The multilayer system can be described as a product of matrix $\prod_{j=1}^x M_j = M = \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix}$ with x number of layers. The characteristic matrix of each layer M_j , with $j=(1,2, \dots,x)$, is:

$$M_j = \begin{bmatrix} \cos(\varphi_j) & -\frac{i}{p_j} \sin(\varphi_j) \\ -ip_j \sin(\varphi_j) & \cos(\varphi_j) \end{bmatrix} \quad (1)$$

where the argument φ_j of the trigonometric functions in the matrix is the phase variation of the light wave passing through the j th layer, and for normal incidence $\varphi_j = (2\pi/\lambda)n_j d_j$, where n_j is the refractive index of the layer and d_j its thickness. The parameter i is employed for transverse electric (TE) wave and, in particular, $p_j = \sqrt{\epsilon_j/\mu_j}$. The parameter q_j , equal to $1/p_j$, replace p_j in the case of transverse magnetic (TM) wave (taking into account that, at normal incidence, the transmission spectra for TE and TM waves are the same).

From the final matrix M we can determine the transmission coefficient

$$t = \frac{2p_s}{(m_{11} + m_{12}p_0)p_s + (m_{21} + m_{22}p_0)} \quad (2)$$

in which p_s is for the substrate and p_0 is for air. Consequently, the transmission:

$$T = \frac{p_0}{p_s} |t|^2 \quad (3)$$

The dispersion of the refractive index for SiO₂ has been taken from Ref. [34]:

$$n_{\text{SiO}_2}^2(\lambda) - 1 = \frac{1.9558\lambda^2}{\lambda^2 - 0.15494^2} + \frac{1.345\lambda^2}{\lambda^2 - 0.0634^2} + \frac{10.41\lambda^2}{\lambda^2 - 27.12^2} \quad (4)$$

While the dispersion of the refractive index of TiO₂ has been taken from Ref. [35]:

$$n_{\text{TiO}_2}(\lambda) = \left(4.99 + \frac{1}{96.6\lambda^{1.1}} + \frac{1}{4.60\lambda^{1.95}} \right)^{1/2} \quad (5)$$

The complex refractive index dispersion has been taken from Refs. [28,29], while the refractive index dispersions for two-dimensional hybrid organic-inorganic perovskite semiconductors (Ruddlesden-Popper (RP) phase and Dion-Jacobson (DJ) phase) have been taken from Ref. [30].

3. Results and discussion

We have designed three different microcavities, characterized by perovskite layers embedded between two periodic photonic crystals, two aperiodic photonic crystals, or two disordered photonic structures. We have employed three different types of perovskite semiconductors: MAPbI₃, monolayered Ruddlesden-Popper (RP) phase, and monolayered Dion-Jacobson (DJ) phase. In particular, the first two are largely under current investigation in the research community and hence an interesting platform for study. Instead, the DJ is an alternative two-dimensional phase that is processed similar to the RP perovskites. We

chose the three types of photonic crystals to highlight the peculiarities of these sequences, which are a very precise bandgap in the case of the periodic one and a deterministic series of valleys and peaks in transmission for the aperiodic case, and random occurrence of transmission peaks in the case of disordered photonic structures.

The periodic microcavity that embeds the perovskite layer consists of a photonic crystal of 10 bilayers of TiO₂ and SiO₂, the layer of MAPbI₃, another photonic crystal of 10 bilayers of TiO₂ and SiO₂ (Fig. 1a). The aperiodic microcavity consists of two Thue-Morse quasicrystals that embed the perovskite layer. Each Thue-Morse crystal is composed of 32 layers with the following sequence: ABBABAABBAABBABAA-BABBAABBABAAB, in which A is TiO₂ and B is SiO₂ [36] (Fig. 1b). The disordered microcavity consists of two disordered photonic structures that embed the perovskite layer. Each disordered structure is composed of 32 layers in which each layer has a 50% probability of being either TiO₂ or SiO₂. The sequence of layers for the first disordered structure is BBABBAABBBABBABAABBBBABBABABABA, while the sequence for the second structure is AAABBABAABBAABBBABBAAABABABABB (Fig. 1c).

For the MAPbI₃-based microcavities, the thicknesses of the layers are 135 nm, 175 nm, and 350 nm, for TiO₂, SiO₂, and MAPbI₃, respectively. In Fig. 2 we show the transmission spectra of microcavities embedding MAPbI₃, with the periodic microcavity in Fig. 2 top, the aperiodic Thue-Morse microcavity in the center, and the disordered microcavity in Fig. 2 bottom.

We have simulated microcavities in which the embedded layer is the Ruddlesden-Popper (RP) phase of a two-dimensional hybrid organic-inorganic perovskite semiconductor. We have selected the monolayer of the perovskite semiconductor, that we call RP1 as in Ref. [30]. In the case of the monolayer, the formula of the compound is (CH₃(CH₂)₃NH₃)₂PbI₂ [30]. In Fig. 3 we show the transmission spectra of a periodic (top), an aperiodic Thue-Morse (center), and a disordered cavity (bottom). For these cavities the thickness of the titanium dioxide layers is 93 nm, the thickness of the silicon dioxide layers is 120 nm, and the thickness of the RP perovskite layer is 350 nm.

Finally, we have simulated microcavities in which the embedded layer is the Dion-Jacobson (DJ) phase of a two-dimensional hybrid organic-inorganic perovskite semiconductor. We have selected the monolayer of the perovskite semiconductor, that we call DJ1 as in Ref. [30]. In the case of the monolayer, the formula of the compound is 4-(aminomethyl)piperidinium PbI₄ [30]. In Fig. 4 we report the transmission spectra of the DJ1 phase embedded in a periodic (top), an aperiodic (center), and a disordered cavity (bottom). The thickness of the layers for these structures with DJ1 is 120 nm, 150 nm, and 350 nm for TiO₂, SiO₂ and DJ1, respectively.

From the results of Figs. 2–4 it becomes clear that the transmission spectra of the 2D perovskites RP and DJ are dominated strongly by the transmission peaks of the photonic structures. The MAPbI₃ absorb up to a wavelength of 750 nm, hence only in the near-infrared part of the spectra the features due to the photonic structures are predominant.

To design a tunable structure we have employed a photochromic polymer, pDTE [37], as an additional layer to induce, upon light irradiation, a shift of the spectral feature of the microcavity. It has been reported that ultraviolet irradiation induces a change from a colourless form of pDTE to a blue form of pDTE [37]. In this case, the microcavity is composed by a 10-bilayer TiO₂-SiO₂ photonic crystal, a layer of pDTE, a layer with the monolayered RP phase, a layer of pDTE, and another 10-bilayer TiO₂-SiO₂ photonic crystal. In Fig. 5 we show the transmission spectra of the cavity in the case of the transparent phase of pDTE (black solid curve) and in the case of the blue phase of pDTE (red dashed curve).

The complex refractive index of pDTE in the two forms has been taken from Ref. [37]. The transmission valley centred at 600 nm for the blue form is due to the strong absorption of the form in this spectral region. We can observe a shift of 15 nm in the cavity mode at longer wavelengths (about 800 nm) due to the photochromic behaviour of

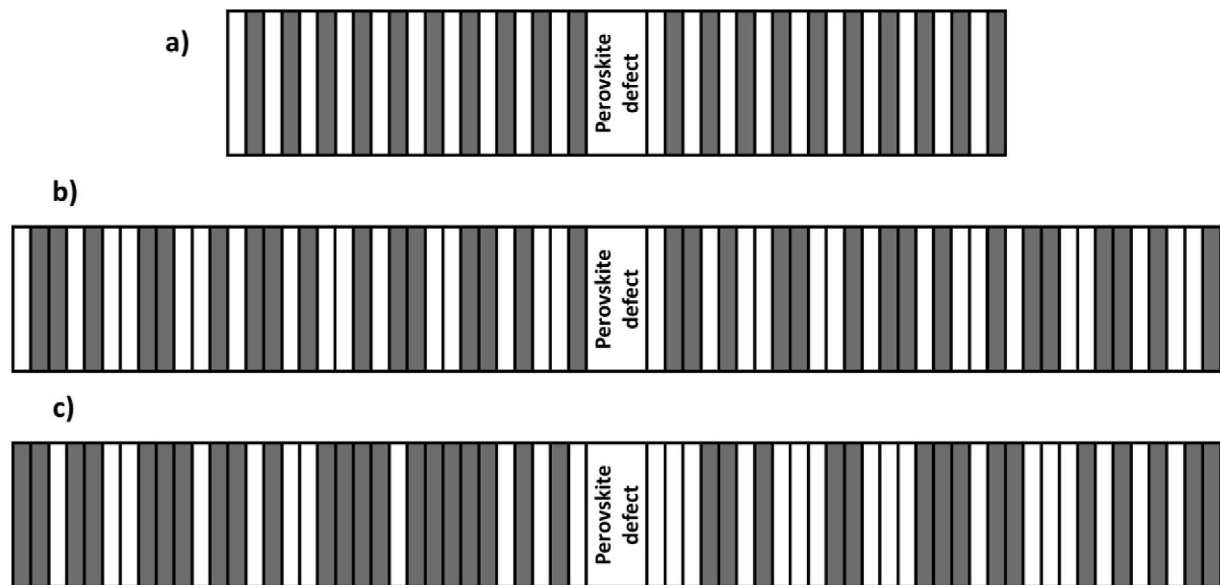


Fig. 1. Sketches of the microcavities with the perovskite layer as defect. a) Periodic microcavity; b) aperiodic microcavity; c) disordered microcavity. The white layer is TiO₂ and the black layer is SiO₂.

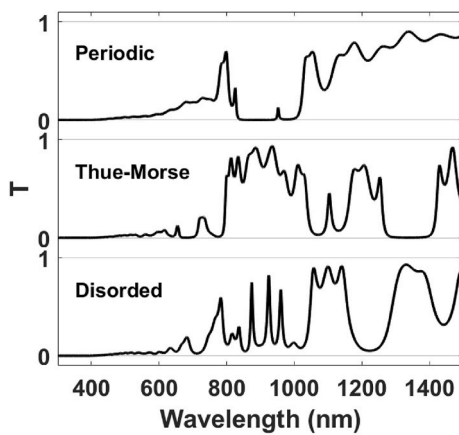


Fig. 2. Transmission spectra of the MAPbI₃ layer embedded in periodic, aperiodic (Thue-Morse), disordered microcavities.

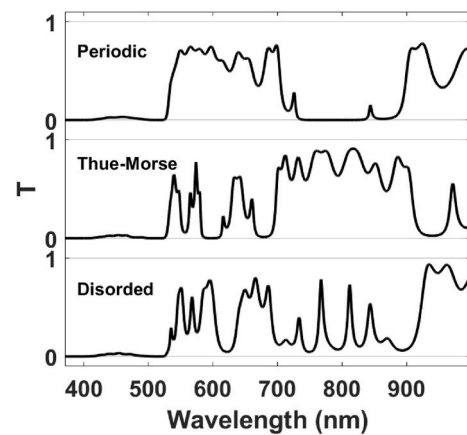


Fig. 4. Transmission spectra of the DJ1 layer embedded in periodic, aperiodic (Thue-Morse), disordered microcavities.

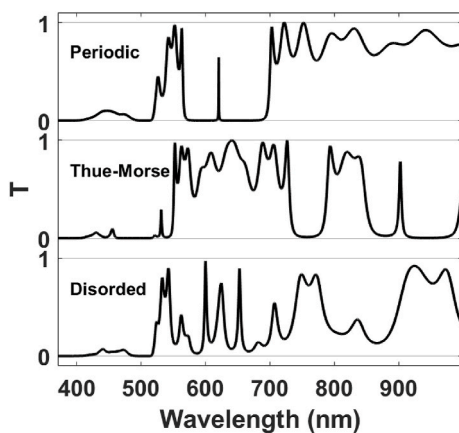


Fig. 3. Transmission spectra of the RP1 layer embedded in periodic, aperiodic (Thue-Morse), disordered microcavities.

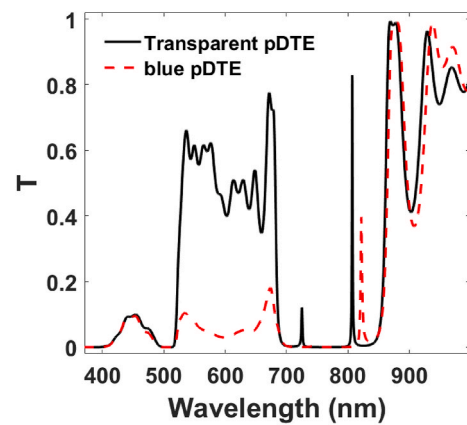


Fig. 5. Transmission spectra of the periodic cavity [(SiO₂/TiO₂)x10]/pDTE/PR1/pDTE/[(SiO₂/TiO₂)x10] with transparent phase of pDTE (black solid curve) and blue phase of pDTE (red dashed curve).

pDTE.

4. Conclusion

To conclude, we have designed a one-dimensional microcavity that embeds MAPbI₃ and 2D hybrid organic-inorganic perovskite semiconductors by using the transfer matrix method. Such active materials have been sandwiched between periodic, aperiodic and disordered photonic structures. To engineer the perovskite layer we have taken into account the complex refractive index dispersion of the materials, recently reported in literature, together with the refractive index dispersions of silicon dioxide and titanium dioxide. In order to conceive a tunable perovskite-based microcavity, we have employed pDTE, which is photochromic materials able to change the refractive index upon ultraviolet irradiation.

CRediT authorship contribution statement

Michele Bellingeri: Conceptualization, Methodology, Validation, Data curation, Writing – review & editing. **Francesco Scotognella:** Methodology, Validation, Data curation, Writing – original draft, Writing – review & editing, Funding acquisition.

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.

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