Control of spontaneous emission rate in lead halide perovskite film on hyperbolic metamaterial

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Abstract. Hyperbolic metamaterials represent a class of nanophotonic architectures with the possibility of controlling density of optical states. Due to this property, hyperbolic metamaterials can be employed as meta-electrodes in optoelectronic devices. On the other hand, lead halide perovskites have several promising properties for application in light-emitting devices. Moreover, a perovskite film is easily deposited on a hyperbolic metamaterial surface. Here, we theoretically show how to accelerate radiative recombination in a perovskite film with a hyperbolic metamaterial. This effect can be applied in light-emitting devices, where radiative recombination is extremely important.

1. Introduction
One of the structures, which supports high density of photonic states is a hyperbolic metamaterial (HMM)\cite{1, 2}. Due to hyperbolic dispersion, these metamaterials maintain states with extremely high wavelength vectors\cite{1} that allows to accelerate spontaneous emission\cite{3}. This effect is useful for such nanophotonic applications as lasing \cite{4, 5} and charge transfer\cite{6}. On the other hand, acceleration of radiative recombination can be vastly useful in lead halide perovskite optoelectronic devices where high quantum yield is a crucial parameter\cite{7}. Moreover, since perovskite emission wavelength is easily tuned\cite{8}, it can be matched to HMM resonance, as it was demonstrated for various nanophotonic designs \cite{9, 10, 11}.

In this work, we present a theoretical study of radiative properties of a dipole source with various orientations located at different position inside a perovskite film on the HMM. We revealed the optimal wavelength at which a significant enhancement of radiative recombination and the maximum available value of the Purcell factor are expected.

2. Results
By definition, HMMs are anisotropic media exhibiting hyperbolic isofrequency contours\cite{1}. For periodic metal-dielectric structures with permittivities $\varepsilon_{m}$, $\varepsilon_{d}$, and thicknesses $t_{m}$, $t_{d}$, respectively, the valid effective medium approximation predicts a uniaxial permittivity tensor with the following components:

\[
\varepsilon_{\parallel} = p\varepsilon_{m} + (1 - p)\varepsilon_{d},
\]

\[
\varepsilon_{\perp} = \left( \frac{p}{\varepsilon_{m}} + \frac{1 - p}{\varepsilon_{d}} \right)^{-1},
\]
where $p = t_m/(t_m + t_d)$ is the metal fill fraction in the unit cell. To achieve the hyperbolic regime, parallel $\varepsilon_\parallel$ and perpendicular $\varepsilon_\perp$ components of the tensor (relatively to the anisotropy axis) must satisfy the condition: $\varepsilon_\parallel \cdot \varepsilon_\perp < 0$.

In our study, we examine the HMM as a multilayered periodic structure with a period of 10 nm gold and 10 nm alumina layers. The fabrication method of this kind of structure is well developed[12]. We assume that $\text{Al}_2\text{O}_3$ permittivity equals to 3.06 and Au has dispersion from[13]. Considering these parameters, our estimation based on Equations 1 and 2 reveals that for a wavelength above 500 nm HMM of type II ($\varepsilon_\parallel < 0, \varepsilon_\perp > 0$) is expected.

Numerical procedure to calculate the Purcell factor was conducted applying the impedance approach[14]. All our calculations were performed with the CST Microwave Studio. The size

**Figure 1.** Schematic model design and theoretical dependence of the Purcell factor on wavelength for emitter located at different distance from HMM surface with parallel (a) and perpendicular (b) orientations.
of a dipole emitter was considered with dimension of about \( \sim \lambda/100 \), that is equal to 3 nm. On the top of the hyperbolic metamaterial with 7 periods a 100 nm thick perovskite film with permittivity of 6.3 was placed. Open boundaries and two symmetry planes were applied to reduce the size of the computational domain. The calculations are made for two different orientations of the dipole: parallel (right Figure 1a) and perpendicular (right Figure 1b) to the surface. The dipole emitter was located in the perovskite film at distance \( d \) from 3 nm to 60 nm from the HMM.

According to our simulations, the spontaneous emission rate of the dipole emitter near the HMM is enhanced. The calculated Purcell factor for parallel and perpendicularly oriented dipoles located at various distances from the HMM are shown in Figures 1a (left) and 1b (left), respectively. For both orientations, the highest values of the Purcell factor is observed at the wavelength of 600 nm for the closest to the HMM emitter. The value of the Purcell factor for a perpendicular to the surface dipole is more than three times larger than for a parallel one. Based on electric field distribution, the coupling between high-k mode and dipole is stronger at this orientation.

With the shifting of the dipole emitter from the HMM surface, the value of the Purcell factor diminished drastically due to decrease of the coupling. At distance more than 20 nm, the value of spontaneous emission rate was practically equal to the value in a perovskite film with the HMM absence. The average value of the Purcell factor can be calculated as an arithmetical mean of the three different dipole orientations. Its highest value equals to 4.8 at 600 nm wavelength for the dipole near the HMM surface. However, due to strong distance dependence, to estimate the Purcell factor in a real perovskite film, averaging over the film thickness is required.

3. conclusion
We have studied Purcell factor in lead halide perovskite film on the hyperbolic metamaterial and revealed that for a perovskite with emission at wavelength 600 nm it is possible to reach acceleration of radiative recombination. Our numerical simulations are relevant for wide range of lead halide perovskite materials APbX\(_3\) (where A is cation, and X is Cl\(^-\), Br\(^-\), I\(^-\) or their mixture), and their agree with recent experimental studies of perovskite emission interaction with HMM [15, 16, 17]. This approach can be applied for creation of efficient light emitting devices [18, 19] and solar cells [7, 20], where acceleration of radiative recombination plays one of the key roles [21].

Acknowledgments
This work was supported by the Russian Science Foundation (project 19-73-30023).

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This work was supported by the Russian Science Foundation (project 19-73-30023).

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